

Small-Scale Carbon Capture Systems

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Carbon capture is the most costly phase of a carbon capture, utilization, and storage (CCUS) supply chain, due to high capture cost from sources, which are diluted in CO₂. This is especially true at a small-scale, as CO₂ content in flue gases are significantly lower than those from large scale energy generation. The values of the cost of CO₂ captured and the cost of CO₂ avoided reported by Rubin et al. would be much lower than those for small-scale generation due to the diluted CO₂.

Keywords: carbon capture (CC) ; carbon capture and storage (CCS) ; micro-combined heat and power (micro-CHP) ; micro-cogeneration ; energy systems ; buildings ; GHG emissions

1. Introduction

The International Energy Agency (IEA) has set a goal of carbon neutrality by 2100 for climate change mitigation. Additionally, the Paris Agreement has set a global initiative to reduce global warming to much less than 2 °C and to further engage efforts to reduce it to 1.5 °C ^{[1][2][3]}. Despite the increasing use of renewable energy sources, their growth is still insufficient to switch from fossil fuel consumption by the end of the century. Further, the current trend will limit the rise in global temperature to 2.7 °C by 2100, which is not sufficient to meet the 2 °C reduction target. Carbon capture, utilization, and storage (CCUS) is a proposed solution to achieve carbon neutrality during the transition from fossil fuels to fully renewable energy generation. Carbon capture (CC) is an attractive option as it aims to achieve carbon neutrality, while simultaneously generating waste carbon dioxide (CO₂) that can be converted into products and sold for profit. Further, decentralized energy generation has been widely investigated as a possible developmental path for achieving carbon neutrality. Distributed power generation in the form of small, decentralized systems can support decrease in emissions and protection of grid capacity, while also offering options for renewable energy ^{[4][5][6][7][8]}.

There exist numerous ones on large-scale carbon dioxide capture, utilization and storage ^{[9][10][11][12][13][14][15][16]}, carbon capture and storage (CCS) ^{[17][18][19][20]}, demonstration and deployment of CCS systems ^{[21][22][23][24][25][26]}, cost of CCS ^{[27][28]}, CCS applied in industry ^{[29][30][31]}, CC ^{[32][33]}, integration of CC in power generation plants ^{[34][35]} and in community scale energy systems ^{[36][37]}, CC in pre-combustion, post-combustion and oxy-combustion in thermal power plants ^[38], CC post-combustion by chemical-looping ^[39], chemical absorption ^{[39][40][41][42][43][44]} or physical adsorption ^{[45][46][47]}, membrane-based CC ^{[48][49][50]}, and, finally, CC and separation technologies for end-of-pipe applications ^[51]. Although there is a myriad of dedicated ones on large-scale CCUS-CCS or CC systems, available ones on small-scale CCS for building applications are very limited.

2. Process and Configurations

2.1. Micro-Gas Turbine

The micro gas turbine (mGT) is a suitable technology for small-scale heat and power generation, given the push towards decentralized heat and power production. For decentralized, small-scale generation to be favorable, the solutions must be both carbon free and efficient. Within this framework, mGTs are an attractive solution as they provide high flexibility, as well as a global efficiency of 80% (electrical efficiency of 30% and thermal efficiency of 50%) ^{[52][53][54][55][56][57]}. The range of electrical power output from a typical mGT is between 50–500 kW, which is suitable for small-scale use in multi-family residential, commercial, and institutional applications. The available one focusses on the Turbec T100 mGT ^[58], with a nominal power output of 100 kW, as it is well-known and can be considered representative of the current state of the art. The typical CC method uses an absorber-stripper system where the absorbent is a 30 wt% aqueous monoethanolamide (MEA) solution ^{[40][42]}.

The Turbec T100 is a typical recuperative Brayton cycle mGT ^{[58][59][60]}.

The general process of the cycle investigated by De Paepe et al. [61] can be described as follows: The air is compressed in a variable speed radial compressor (1) and passed through a recuperator (2) where it is preheated by the exhaust gas arriving from the turbine. The preheated air then enters the combustion chamber, where burning natural gas heats it to a nominal outlet temperature of 950 °C (3). The combustion gas mixture expands across the turbine (4), which delivers the power to drive the compressor and converts the excess power to electricity through a variable speed generator (5). The heat remaining in the gas after the recuperator is recovered through a gas-water heat exchanger (6), which can be used to heat water for combined heat and power systems. The exhaust gases are then split into two streams, one of which is recirculated to the compressor, and the other goes to the CC plant. Exhaust gas recirculation is one of the known emission control technologies for reducing NO_x emissions by recirculating a part of the exhaust gas, while reducing fuel consumption and pumping loss. The EGR ratio is defined by the ratio of intake CO₂ concentration to exhaust CO₂ concentration.

The EGR stream passes through a cooler (7) to maintain high compression efficiency (to be discussed in a later section), and the condensed water is separated (8). The gas is next distributed over a blower (9) to provide a driving pressure increase, followed by a filter (10), which leads to the compressor inlet. A certain ratio of exhaust gases is also directed to the CC plant. The CC plant has two columns, one absorber and one stripper. A blower (11) provides the required pressure to drive the flue gas in the bottom of the absorber (13). The lean solvent is fed into the top. The interaction of the gas and liquid phases in the absorber drives the CO₂ to the liquid phase, as a result of the concentration gradient at the liquid/gas interface. The rich solvent is then pumped through the heat exchanger (15), where it is heated to a higher temperature by the lean-solvent from the stripper bottom. It then enters the stripper column, where the solvent is regenerated through heat provided by pressurized hot water (16). The vapor at the top of the desorber enters the condenser, in which the water is removed, and nearly pure CO₂ is obtained. The regenerated solvent is pumped back through the rich-lean heat exchanger, and cooled further by an air-cooled plate cooler (18). Wash columns are also mounted at the top of the stripper and absorber columns to eliminate entrained droplets of solvent transported by the flue gas by means of demineralized water. Wash columns have a low energy effect on the CC plant and are neglected in the numerical analysis performed by Giorgetti et al. [59][60].

This configuration, with the PACT amine capture plant combined with a Turbec T100 mGT is considered by Akram et al. [62], Majoumerd et al. [63] and Ali et al. [64][65]. Their individual findings are detailed in the following performance section of the paper.

Due to the low CO₂ concentration (~1.5 vol%) and high volumetric flowrate of exhaust gases that enter the CC plant, in addition to the substantial residual O₂ amount due to CC plant integration, several cycle modifications have been proposed. One such modification is selective EGR (S-EGR), as opposed to the traditional EGR cycle. Bellas et al. [66] examined the influences of the S-EGR cycle on the mGT cycle for CC applications. The S-EGR system is similar to the traditional EGR cycle; however, the separated flue gases are passed through a selective membrane system that uses an air sweep stream that blends with the CO₂ passing over the membrane. The CO₂ and air are recirculated to the compressor inlet, while the CO₂ exhausted gases are released to the atmosphere. The parallel and series configurations investigated by Bellas et al. [66].

However, the most common humidification cycle is the micro humidified air turbine (mHAT) cycle. Giorgetti et al. [59] assessed the performance of the mHAT cycle through modification of the standard mGT-EGR cycle through the addition of a saturation tower. This cycle can be considered typical throughout the examined one. The general cycle process is modified as follows: After passing through the compressor the air is humidified at the saturation tower (11). In this configuration, the gas-water heat exchanger (6) is used to heat up the water for the saturation tower, and is no longer, or only partly, used for cogeneration purposes. To balance the water humidification, feedwater enters the circuit (12). A variable pressure pump is also added to drive the circulation of water (13).

Majoumerd et al. [63] assessed the performance of a typical mGT and mHAT coupled with an advanced post-combustion CO₂ capture unit. The SOA chemical absorption unit also implements monoethanolamine (30 wt% MEA) as a chemical solvent. The capture plant configuration is similar to that studied by Giorgetti et al. [59], but the additional cooler for the lean solvent is removed.

2.2. Hybrid Fuel Cell Systems

Fuel cells such as those in small-scale applications [67][68] or in micro-scale applications [69][70], have high efficiency, as electricity is generated through an electrochemical reaction as opposed to a series of energy conversions. Hybrid fuel cell systems on a small-scale are scarce, while hybrid systems with large-scale gas turbines are more readily available, such

as hybrid solid oxide fuel cells directly coupled to a gas turbine [71][72], or indirectly coupled to a gas turbine [73] in power plants, and hybrid solid oxide fuel cell-gas turbine cycles using alternative fuels [74].

Roohani Isfahani and Sedaghat [75] developed a novel system with a unique combination of a solid-state fuel cell, micro gas turbine, and CC unit to use natural gas energy in a more effective approach. The fuel cell power output considered was between 950 and 1360 kW, making the system applicable to decentralized energy generation with application to multi-use commercial buildings, as opposed to single-family dwellings and small businesses. The system comprises three reactors for splitting hydrogen and carbon dioxide from natural gas through the three-reactor chemical looping hydrogen generation (TRCL). Several others are available that focus on hybrid power plants of solid oxide fuel cell (SOFC), and micro-GTs operating at baseload [76][77] or part-load [78], applying various operating strategies [79][80] or using alternative fuels [81] for micro-CHP applications. However, Roohani Isfahani and Sedaghat [75] extended the above systems to integrate with a CC plant and TRCL system, which is pertinent to the topic. The hybrid power plant is made up of a reformer, three-reactor chemical looping for hydrogen production, a fuel cell, micro-GT, an internal heat exchanger, and the CO₂ capture loop. Natural gas is broken down into carbon dioxide and hydrogen constituents in the reformer, which is then passed into the TRCL reactors. The TRCL comprises of three reactors: fuel, steam and air reactors. In the first stage, fuel is injected into the fuel reactor (FR). In this reactor, hematite (Fe₂O₃), which contains a significant amount of elemental oxygen, is mainly reduced to FeO (Wüstite, a mineral form of iron (II) oxide) at 950 °C at the upper riser, and 890 °C at the bubbling bed. The fuel is converted into CO₂ and H₂O. In this stage, the CO₂ is ready to be absorbed as a product. In the second reactor (steam reactor or SR) FeO (Wüstite) reacts exothermically with steam and creates magnetite (Fe₃O₄) and hydrogen at 950 °C. In the air reactor (AR), entered magnetite (Fe₃O₄) reacts exothermically with pure air, to produce hematite (Fe₂O₃) and oxygen-depleted air as products. The overall reaction changes methane (CH₄) into hydrogen and CO₂. CH₄ is also converted to H₂ and CO₂. Therefore, CO₂ is integrally separated from fuel. The TRCL reactors are operated at a pressure of 3 bar.

Steam from the heat recovery steam generator reacts with fuel in the reformer. The H₂ from the steam reactor is then fed into the anode of the fuel cell, and O₂ depleted air from the air reactor goes to the cathode to start electricity production. The fuel reactor exhaust CO₂ is fed to the CO₂ capture loop. Unreacted hydrogen from the SOFC is fed into the combustor and burned to produce sufficient hot gas to run the micro gas turbine. This latter produces energy that runs the adjacent compressor to compress the inlet air to the air reactor. The turbine exhaust gases are also used to preheat the incoming natural gas. In the CO₂ capture loop, the incoming CO₂ from the fuel reactor of the TRCL is cooled in the CO₂ heat recovery steam generator (CO₂ HRSG). The CO₂ undergoes several compression and cooling processes to reach the pressure required in the pipeline.

2.3. Biomass-Fired Organic Rankine Cycle

The organic Rankine cycle (ORC) system is an attractive technology for cogeneration applications in the 200–1500 kW range, mainly used in waste heat recovery [82][83][84], at small-scale for industrial or commercial buildings [85][86], in small-scale and micro-scale biomass fueled CHP systems [87], or at micro-scale for residential applications [88]. However, few applications with small-scale ORCs with integrated CCS are commercially available. Zhu et al. [89] designed a biomass-fired ORC for small-scale CHP systems, coupled with an MEA-based CC unit, for which they performed an extensive thermo-economic simulation one. The system implements biomass combustion as the primary energy source and the ORC as secondary. The electric power output for the considered system ranges from 100–500 kW and is therefore applicable for small-scale applications, such as residential and commercial buildings, office building blocks and so on. The process is defined in the following manner: The biomass fuel is combusted in the biomass boiler and heat is transferred to the pressurized hot water during process 9–8. The heat is then absorbed in the evaporator (4–1) by the organic working fluid, which then passes over the expander to produce power (1–2). The working fluid then enters the condenser where it is cooled by the cooling water (2–3). The cooling water absorbs the waste heat (10–11) to generate domestic hot water or discharges the heat in the cooling towers (17–18). From the biomass boiler, the flue gases preheat the combustion air entering the boiler (7–16), and then enter the MEA chemical absorption unit.

3. Performance

3.1. Micro Gas Turbine

Micro gas turbines offer the lowest CO₂ emissions per produced kW, making them an attractive option for small-scale CHP energy generation systems. However, the carbon dioxide emissions must be captured in order to achieve carbon neutral energy production. Challenges encountered when integrating carbon capture technology into mGT energy conversion cycles result most notably from the low CO₂ concentration and high volumetric flow rate of exhaust gases

(~1.5 vol%), plus the significant remaining O₂ concentration that leads to solvent degradation. These factors have a negative impact on the size, energy use and economic performance of the downstream CC plant, while also resulting in solvent degeneration if an amine-based CC plant is utilized. In order to minimize these effects, namely the cost of the energy penalty, technologies such as auxiliary firing, exhaust gas recirculation (EGR), selective exhaust gas recirculation (S-EGR), humidified cycles, and oxy-fired gas turbines cycles have been proposed. Supplementary firing and oxy-fired GT cycles have not been explored on the small-scale one; therefore, the following subsections detail the overall impact of the CC plant, followed by the impact of EGR, S-EGR, and humidification on cycle performance.

3.1.1. Impact of Carbon Capture

As previously described, integration of a CC plant with an mGT results in a high energy penalty. Giorgetti et al. [60] assessed the effect of a CC plant on the global performance of the mGT through numerical simulations in Aspen Plus. Their results indicated that the cycle performance was greatly impacted by the thermal energy demand for the stripping process (reboiler duty), reducing the total electric efficiency by about 6.2 absolute percentage points

It is also important to point out the influence of ambient air temperature on electrical efficiency. As ambient air temperature rises, the air density reduces, causing a lower mass flow of air within the engine and reduced power production. Consequently, a higher heat input is required to increase the air and fuel mass flow to generate the nominal power output, resulting in efficiency decrease. The ambient air temperature also affects the oxygen concentration at the combustor inlet; however, these effects are marginal and are not of concern [63].

3.1.2. Exhaust Gas Recirculation

Exhaust gas recirculation (EGR) is a proposed technology to decrease the cost of the energy penalty from CC resulting from the low CO₂ content of exhaust gases. It is worth noting that post-combustion CC is more efficient for large-scale GTs where CO₂ concentrations are higher (~3.8 to 4.4 mol%) than for mGTs (~1.6 to 1.8 mol%) [64]. Exhaust gas recirculation supplies three main benefits—it increases the exhaust gas CO₂ concentration for a reduced carbon capture energy penalty, it reduces harmful NO_x emissions, and it decreases the volumetric flow rate to the CC plant by recirculating a fraction of the exhaust gases back to the compressor inlet [61]. Akram et al. [62] experimentally determined that, per unit percentage rise in CO₂ concentration, the specific reboiler duty decreased by around 7.1%, and numerically predicted a 6.6% reduction.

However, EGR also introduces some challenges. The combustion stability diminishes and unburned emissions increase as the O₂ concentration at the combustor inlet is decreased; electrical efficiency is decreased through auxiliary energy losses to the fan that drives the recirculated gas, and the compressor inlet temperature is increased, resulting in a slight decrease in thermodynamic performance [90]. Giorgetti et al. [59][90] found that the EGR blower consumed 4.5 kW of power at nominal operation conditions accounting for 4.5% of the electrical power output. Removing this from the efficiency calculation resulted in nearly identical efficiency relative to the traditional mGT cycle. The remaining difference was small due to the change in the inlet mixture temperature with EGR.

Majoumerd et al. [91] found that with 40% EGR the CO₂ content of the exhaust gas could be increased from 2 mol% to 3.4 mol%, through simulations with a validated thermodynamic model. This represents a 67% increase relative to the reference mGT cycle. Similarly, through simulations using Aspen Hysys and IPSEpro, Ali et al. [64][65] established that the CO₂ concentration in the exhaust gas of the mGT with 55% EGR was 2.2 times greater than the traditional mGT cycle, where baseline CO₂ content in the exhaust was 1.46 mol%. This increase resulted in a 40% reduction in specific reboiler duty, demonstrating the advantages of EGR for CC applications, owing to the reduction in cost from smaller absorption/stripping columns and the reboiler.

Majoumerd et al. [63] determined that while the CO₂ concentration in flue gases was increased with EGR, the global efficiency was decreased from 23.0% without EGR to 22.5% with EGR. Ali et al. [64][65] came to the same conclusion, that electrical efficiency of the mGT cycle is decreased by EGR. Their results showed a decrease in mGT electrical efficiency from 32.1% to 29% at a 55% EGR ratio, when the effects of the CC plant were not considered. As discussed, this can be attributed to the blower power required for recirculation and the changes in the fluid thermodynamic properties that effect compressor and turbine operation. However, the global efficiency reduction due to EGR is small, and it appears unanimous in the conclusion that EGR is desirable due to the cost decrease for CO₂ capture.

Best et al. [92] experimentally assessed the effects of EGR on mGTs through CO₂ injection in a Turbec T100. Their results showed that, at low power outputs (50 kW) with 125 kg/s CO₂ augmentation, CO emissions increased by 109% and unburnt CH₄ emissions by 338%. However, they concluded that emissions were not significantly impacted at higher load

factors. Further, due to the lower combustion temperatures, NO_x emissions showed a decreasing trend with CO_2 enhancement.

3.1.3. Selective Exhaust Gas Recirculation

As discussed, EGR increases the CO_2 concentrations of exhaust gases while decreasing the volumetric flow rate to the CC plant by recirculating a portion of the exhaust back to the compressor inlet. This has been demonstrated to reduce the energy penalties of carbon capture, as well as to decrease the capital cost of the system as a result of the reduced system size. However, increasing the fraction of flue gas to be recirculated decreases the oxygen concentration at the combustor inlet, resulting in flame instabilities and decreased combustion efficiency, and high CO and unburned hydrocarbon (UHC) emissions. Therefore, the EGR ratio is limited by an optimal O_2 concentration of 16 vol% at the combustor inlet. S-EGR is a proposed cycle modification to increase CO_2 content of exhaust gases to a higher degree than achievable with EGR, without compromising O_2 concentrations at the combustor inlet required for combustion stability.

Darabkhani et al. [93] researched the performance of a parallel S-EGR configuration through both simulation and experimental testing. The focus was on the performance of a commercially available, polydimethylsiloxane organic polymer membrane (purchased from PermSelect Ltd., Ann Arbor, MI, USA), as investigated both experimentally and through simulation. Through process simulations, it was found that CO_2 concentrations could be achieved of up to 14.9% with 60% EGR, with a 90% CO_2 removal rate from the membranes.

Challenges encountered with S-EGR are flame instability and combustion efficiency leading to increased CO and UHC emissions due to the higher CO_2 content achieved in the flue gases. Bellas et al. [66] performed experimental ones on the Turbec T100 mGT with the goal of investigating the effects of CO_2 enrichment on the performance of an mGT. To emulate the effects of S-EGR, CO_2 was injected into the compressor inlet. Injection rates of 0 to 300 kg/h CO_2 (1.7 to 8.4 vol% at 100 kW and 1.4 to 10.1 vol% at 60 kW) were considered, and the effects on gas turbine performance were assessed. This represents a nearly six-fold increase in CO_2 concentration, which is typical of S-EGR. It was found that high levels of CO_2 injection modified the specific heat capacity and density of the oxidizer, decreasing the engine speed and system temperatures. The CO and UHC emissions increased greatly at part loads, whereas at nominal load they experienced little change with increased injection rates. This is an effect of incomplete combustion caused by poor fuel and air mixing, inadequate flame stability, and lesser combustion temperatures. At the highest injection rates (300 kg/h of CO_2), the NO_x emissions were lower than at the baseline (no injection). This is a result of lower combustion temperatures at increased CO_2 content.

3.1.4. Humidification

EGR enhances the efficiency of the CC plant, but decreases the electrical efficiency of the mGT cycle resulting in a marginal decrease in global efficiency. Humidification is a proposed method for improving mGT cycle efficiency. In general, the overall economic performance of the mGT powered mCHP system can be improved through improving the electric efficiency of the mGT at the time of low heat demand.

It is available that assess the impact of humidification on the global efficiency of the mGT and coupled CC plant, as well as ones that examine the impact on individual mGT component performance. Both aspects are reviewed in the following discussion.

Giorgetti et al. [59] found that humidification of the traditional mGT cycle can completely offset the efficiency losses introduced by the EGR energy penalty.

Similar results were presented by Majoumerd et al. [63] through simulations using a validated thermodynamic model. The results showed significantly increased performance for the mHAT cycle compared to the traditional mGT, with 25.8% cycle efficiency compared to 23.0% and 22.5% efficiency for the baseline mGT and mGT-EGR cycles, respectively. Of note is that the cycle efficiency includes the effects of the coupled CC plant.

Carrerro et al. [94] experimentally researched the effects of water injection with a saturation tower on the mGT cycle. They found that, overall, the electrical efficiency of the humidified cycle increased up to 4.2 absolute percentage points, similar to that found numerically.

Therefore, it can be concluded that global cycle efficiency is improved through humidification, and entirely compensates for energy losses from EGR. It is important to mention that this result is independent of energy integration between the CC unit and mGT/mHAT, in which waste heat is leveraged to decrease the thermal energy requirement of the stripper. This is

discussed further in the optimization section of this review, in which Giorgetti et al. [59] assess a mGT/mHAT coupled with a CC unit with energy integration between the systems.

3.2. Hybrid Fuel Cell Systems

Roohani Isfahani and Sedaghat [75] evaluated the performance of the hybrid system of a solid-state fuel cell and micro gas turbine with a coupled carbon capture plant. A major benefit of this system is the 100% capture ratio achieved by compressing CO₂ to the liquid state, compared to the 90% CO₂ capture common in the micro gas turbine power generation systems. The influence of plant pressure and SOFC temperature on the system effectiveness and performance has been investigated in detail. The system efficiency can be maximized by increasing the SOFC temperature and operating pressure, although the effect of working temperature clearly has less influence than that of the pressure. It was found that at an SOFC temperature of 1000 °C, an SOFC pressure of 17.5 bar, and a fuel utilization factor of 0.8, the global efficiency reached 48.3% natural gas LHV (lower heating value). It is also important to note that SOFC operating temperatures may not exceed 1000 °C, and that while increasing pressure increases efficiency, it simultaneously increases capital cost, which must be taken into consideration. Moreover, the effect of the utilization factor and plant pressure on power plant efficiency and fuel cell power output has been assessed. The utilization factor may be expressed as the ratio of consumed fuel in fuel cell stacks and the total quantity of fuel introduced to any type of fuel cell and has a typical range of 0.75–0.9. If it is increased, the power output of the fuel cell is increased while the generated power of the gas turbine is decreased. However, given that fuel cells are more efficient than gas turbines, more fuel is utilized in a more effective manner. Therefore, increasing the utilization factor increases the overall efficiency. Further, the maximum net efficiency was found to be 51.4% LHV at an exhaust pressure of 2.5 bar, temperature of 1000 °C, plant pressure of 22.5 bar, fuel utilization factor of 0.9, and CO₂ expanded exhaust pressure of 2.8 bar.

3.3. Organic Rankine Cycle

The biomass-fired organic Rankine cycle is a suitable technology for distributed CHP. Although the ORC is an established technology for cogeneration across the range of 200–1500 kW, few are commercially available on the small- and micro-scale, where the electrical output ranges from 100 to 500 kW. Zhu et al. [89] investigated the ORC-based biomass fueled micro-CHP system with integrated MEA-based CC to assess the thermodynamic and economic performance of eleven working fluids. From the perspective of the feasibility for distributed energy generation below 500 kW, the net power outputs ranged from 175.49 kW for isobutene to 413.82 kW for cyclopentane. Of all the working fluids considered, it was found that cyclopentane had the best thermodynamic performance with a power efficiency of 13.70% and exergy efficiency of 16.21%. This was followed by R141b, R113, R123 and pentane. It is important to note that the reported values in the thermodynamic analysis did not include CO₂ capture. However, from the economic assessment, HFE7000 had the largest net present value (NPV) of 2052.42×10^4 USD and the highest profit ratio of investment (PRI) of 5.45, followed by R1233zd-I, isobutane, isopentane and R113.

4. Overview of Available CCUS Technologies

Several storage options are being considered for carbon capture applications, with varying levels of development. Injecting CO₂ into geologic formations could be considered as it is a mature technology already in use in the oil and gas industry. The main concern associated with geological storage is leakage of the concentrated CO₂ stream and associated environmental damage. However, annual leakage rates range from 0.00001% to 1%. The three main types of geological formation for carbon storage are depleted oil and gas reserves, deep saline reservoirs, and unmineable coal seams [95] [96]. The captured CO₂ can also be sold for profit, which is one avenue to be explored in the interest of offsetting the high costs of CCS. Examples of utilization are direct utilization, enhanced oil recovery (EOR), carbonation, and conversion into chemicals and fuels. Direct uses include the food and drink industry, such as for drink carbonation, the decaffeination process and so on. CO₂ can also be used directly for pharmaceutical applications. Direct utilization is only possible for sources with high purity CO₂ waste streams, such as ammonia production. EOR is the process of using CO₂ to extract crude oil from an oil field or natural gas from unmineable coal deposits. As applied to natural gas, this process is still under development and not yet commercially available. However, EOR for crude oil extraction has been used for many years already in both Canada and the USA. For EOR, CO₂ is injected into otherwise unrecoverable oil reserves for increased oil extraction. Most of the CO₂ is pumped back to the surface and recycled, although some is released into the atmosphere. Under certain conditions, the CO₂ injected for EOR could remain underground as in geological storage. As mentioned, captured CO₂ can also be transformed into fuels and useful chemicals, or used as feedstock for fuel production. Unfortunately, using CO₂ for feedstock results in a highly energy intensive process. Further, fuels and chemicals have a short lifespan, and are quickly released back into the atmosphere. Therefore, the benefits of capture are undermined [21][28][35].

For perspective, the following will provide a brief overview of the large-scale CC projects currently in operation. CCS technology functioning at large-scale has existed since the 1970s. Government subsidized projects to aid develop and commercialize CC plants have been principally concentrated on electricity generation. The major part of CO₂ injection from CC is in EOR in the USA that offers extra revenue to partially offset capture costs. In Canada, there are three capture plants in operation and two in construction as of 2019. The Boundary Dam Carbon Capture and Storage facility in Saskatchewan has been in operation since 2014 with a capture capacity of one million tons of CO₂ on an annual basis [17][88]. The post-combustion capture facility is coupled to a fossil-fueled electricity generating plant, to which CO₂ is largely transported by pipeline and used for EOR at the Weyburn Oil Unit. The remaining CO₂ is transported, also via pipeline, to the nearby Aquistore project for geological storage [15]. The Petra Nova plant in Texas is another commercial large-scale fossil-fueled power plant integrated with CCS technology, with annual CO₂ capture of one million tons. Both plants sell captured CO₂ for EOR, which partially offsets the cost introduced by CCS [97][98][99].

The micro cogeneration systems with integrated CC discussed thus far are purely in the development stage and are not yet commercially available. For instance, Clean O₂ Carbon Capture has developed the first commercial unit for decentralized CC applications [100]. The product provides direct capture to by-product utilization with minimal processing. The process is described as follows: A portion of the flue gases, having a CO₂ concentration of 40,000 ppm or greater, pass through the reaction chamber where caustic soda reacts with the carbon dioxide to create soda ash and water. The flue gases then pass through the reaction chamber into the heat exchanger where waste heat is recovered by heating the municipal water supply for domestic hot water. The caustic soda must be replenished weekly. The Clean O₂ Carbon Capture unit is currently installed in concentrated residential, commercial, and single residential applications. Depending on the use, either a residential or commercial unit may be installed. For concentrated residential applications, a commercial pilot is installed at Garrison Woods and Marda Loop in Calgary. For commercial use, a commercial unit is installed at Westjet airlines in Calgary. Both are projected to produce 6.5 tonnes of by-product, the equivalent of 3 metric tonnes of captured CO₂ per year. In terms of single residential use, a residential pilot is installed in Calgary and projected to produce 630 kg of by-product, the equivalent of 320 kg of captured CO₂ per year. The cost per tonne of CO₂ captured is approximately \$14.94. However, by selling the soda ash by-product, the cost of capture is actually negative. The advantages of using the Clean O₂ Carbon Capture unit are notable, comprising savings of up to 20% on energy charges per annum, and, reflecting ecological concerns, inhibition of GHG emissions into the atmosphere [100].

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