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ENERGY INPUT AND PROCESS FLOW FOR CARBON CAPTURE AND

STORAGE

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ABSTRACT

Carbon dioxide (CO_2) is a primary contributor to global climate change. Efforts to curb climate change include the capture and storage from this carbon, as well as the conversion of carbon gas into clean fuels. Carbon capture and storage (CCS) is a commercially developing technology to capture CO_2 from power generation plants, compress it, and store it in a geologic reservoir. The three main CCS systems (post-combustion capture, pre-combustion capture, and oxyfuel technologies) were compared in terms of carbon capture ability and process flow diagrams were created. From analysis all methods have similar capturing abilities, but oxyfuel technology requires the lowest energy to capture CO_2 and has the lowest CO_2 cost breakeven point. Additionally, other developing carbon capture and utilization methods were discussed, including a gas fermentation process to produce ethanol and direct air carbon capture.

KEYWORDS: Carbon Capture and Storage, Pre-Combustion Capture, Post-Combustion Capture, Integrated Gasification Combined Cycle, Direct Air Capture, Lanza Tech, Gas Fermentation.

1. INTRODUCTION

Greenhouse gases are a primary contributor to climate change, which is a growing concern worldwide. Most of these greenhouse gas emissions, approximately 83% in the United States, are produced from fossil fuels from combustion and nonfuel uses [1]. A primary concern of these emissions comes from anthropogenic carbon dioxide (CO₂). New technologies are constantly being developed in attempt to lower greenhouse gas emissions and slow climate change. One promising development, currently used and constantly being improved, is the use of carbon capture and storage (CCS) technology.

CCS is primarily targeted at electricity generation plants. This is not a trivial sector of the global CO_2 emission sector. Around 1/3 of United States anthropogenic (CO₂) emissions come from power plants [1]. These power plants are built in large centralized units operating at 500-1000 MW of electrical power [2]. A 1000 MW pulverized coal power plant can emit between 6-8 Mt/y of CO₂ [2]. CCS can capture over 90% of the CO₂ released from these large-scale power plants and store it somewhere underground – such as geological sinks or deep in the ocean [3].

There are a few primary reasons CCS is being retrofitted to old power plants and designed connected to new plants. The first, as mentioned, is global climate change concerns. Given the high emission rates of the power production sector, the potentials CCS offers for CO_2 reduction are vast [4]. Additionally, this is one way large emission reductions can be achieved, given the realization that renewable resources will take a considerable amount of time to be implemented on as large of a scale as fossil fuels [4]. Finally, studies indicate CCS will a cost-effective strategy for emission reductions by 2030 and beyond [4].

The process for carbon capture and storage can be described in three simple steps: CO_2 capture and compression, transportation, and storage. CO_2 capture is currently used in a 22 large scale power plants, with 16 in the United States [5]. There are also wide variety of plans in place to retrofit CCS systems to more power plants and build them on new plants [4]. Relatively high capital costs of capture and compression (transport and storage only make up a minor fraction of the total cost) have remained problematic for many power plants [4].

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CCS activities also include geological assessments of CO_2 in storage reservoirs and total environmental impacts [6].

There are three main concepts for carbon capture: post-combustion technology, pre-combustion, and oxy-fuel combustion systems. Post-combustion, the most developed system and currently retrofitted on a few plants, capture typically applies to coal-fuel power generators that are air fired. Pre-combustion works well with gasification plants. Oxy-fuel systems, the least developed concept of the three, can be applied to new power plants [1]. Transport and storage are identical for all methods, as all output compressed CO_2 that is stored in some chosen location. There are also some alternative developments to reduce carbon in the atmosphere, including alternative fuel (ethanol) production from syngas fermentation and direct air carbon capture. This paper will describe the processes for carbon capture as well as energy and material flows for power plants of nominal size for each of the capture methods. It will also describe the alternative technologies developing beyond power plant carbon capture.

2. CARBON CAPTURE PROCESSES FOR POWER GENERATING PLANTS

Post-Combustion Capture

The first part of the CCS process is CO_2 capture and compression. One method for carbon capture is postcombustion capture (PCC). Figure 1 [1,4,6] shows the basic PCC process, typically applied to conventional pulverized coal fuel power plants [7]. In this process, CO_2 is captured from scrubbing the flue gas (the gas after combustion occurs) for CO_2 removal [7]. This method is commercially available and can be retrofit to an existing power plant.

As shown in Figure 1, fuel (coal) and air move through the power cycle as normal - through a boiler to combust and send heat to water to produce power through a steam turbine. After this, the flue gas becomes a mixture of nitrogen and carbon dioxide. The carbon dioxide is at relatively low concentrations which requires special techniques to separate. The flue gas goes through an air pollution control system to separate out some chemicals before going to the carbon capture system.

PCC typically uses chemical absorption in the carbon capture system. This refers to the process in which the CO_2 gas is absorbed in a liquid solvent by forming a chemically bonded compound [2]. For this process, the flue gas passes through the solvent in a packed absorber column where the CO_2 is preferentially removed from the flue gas. Then the solvent passes through a regenerator where the CO_2 is stripped from the solvent by counter flowing steam (100-120 °C). The water vapor is condensed and leaves highly concentrated CO_2 , which is compressed for transport and storage. The solvent is typically cooled and recycled back into the capture system [2].

The most common solvent used in this carbon capture system is monoethanolamine, typically referred to as amines or MEA. Amines react with CO_2 to form water soluble compounds and are capable of capturing the carbon dioxide from streams with low CO_2 partial pressures, such as the flue gas from this pulverized coal power plant [1]. MEA based solvents have high energy requirements, which can be costly and lower the total electrical output of the power plant (as energy is diverted to this system) [7]. Research is currently being done to develop lower energy requiring solvents that also have better thermal integration with the power plant [7].

Though MEA is by far the most used CO_2 capture method in PCC system, development is being done for other systems. Carbonate systems utilize soluble carbonates to react with CO_2 to form a bicarbonate that can be heated to release CO_2 [1]. These have the advantage of lower energy required for regeneration when compared to amine-based systems. Another solvent is ammonia, which operates similarly to amine systems [1]. Membranes have also been considered to separate CO_2 from flue gas [1].







Pre-Combustion Capture

A second method for carbon capture is pre-combustion which is typically called integrated gasification combined cycle (IGCC) carbon capture. This method is at a lower technological readiness level, as it is only now being tested to scale as opposed to commercial availability [7]. IGCC uses gasification, CO_2 capture and H_2 separation, which has the advantage of additional heat converted to power from the hydrogen [7]. Pre-combustion can also be utilized in power plants where natural gas is the primary fuel [2].

A diagram of the basic IGCC process is shown in Figure 2 [1,4,6]. In this process, an air separation unit releases oxygen for gasification with coal/fuel to produce synthesis gas. Note that the air separation unit takes a relatively large amount of energy to work and is expensive. The synthesis gas is mainly a combination of CO and H₂. A shift reactor is used to convert CO to CO₂ with steam (CO + H₂O \rightarrow CO₂ + H₂), producing additional H₂. After this, the H₂ is separated from the CO₂ and the CO₂ is captured. There are several capture techniques, such as methyl diethanolamine (MDEA), or physical solvents (no chemical reactions) such on Rectisol or Selexol [1]. Note that the physical solvents are effective in this approach because the concentration of CO₂ is higher than in PCC. The separated CO₂ is compressed in a similar manner as the PCC process and sent for transport and storage. The H₂ produced during gasification and shift reactor can be sent to a combustion turbine to produce power. Exhaust gas from the combustion turbine can also heat water to run a steam turbine and produce additional power [1].

An alternative to gasification is chemical looping, which has the advantage of producing concentrated CO_2 without the need for a costly air separation unit [1]. For chemical looping, oxygen is supplied by a solid rather than gas oxygen carrier [1]. Both pre-combustion and chemical looping are in low technological readiness levels, with only scaled experiments in place [1].

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Carbon capture and storage - pre-combustion/integrated gasification combined cycle (IGCC) process [1,4,6].

Oxyfuel Carbon Capture

Finally, the last capture method, the least developed system is oxy-fuel combustion (Oxyf). This method uses the combustion of oxygen instead of air, diluted with an external recycle flue gas to lower combustion temperatures and carry energy [7]. This technology has the lowest technological readiness level of all previous methods, at pilot-scale type of studies [7]. Oxy-fuel combustion techniques can be fit onto new power plants involving combustion (not gasification). An advantage to this method is that it has lower SO_x and NO_x than other CCS technologies, and NO is reduced and reburned as it is recycled through [7]. The primary advantage over a system like PCC or IGCC, however, is that the flue gas becomes much more concentrated with CO_2 (almost purely CO_2 and H_2O), meaning the energy and capital costs of chemical or physical separation are not necessary, as water vapor can be easily condensed [2]. However, air separation units again use intensive amounts of power (up to 15% of the plant's electrical output), requiring an increase in fossil fuel use to keep the plant operating at the same power levels [2].

The basic process for Oxyf is shown in Figure 3 [1,4,6]. First, in a similar matter as IGCC, air is separated into its components. The oxygen is combusted with a fuel (such as pulverized coal) in a boiler, producing power with steam turbines. The flue gas from this process goes through a system to remove some contaminants and is then recycled back into the boiler. The flue gas is recycled in order to maintain combustion conditions similar toair fired configurations (to keep the temperature low enough for boiler materials) [1]. Nonrecycled flue gas goes through a CO_2 capture system that only involves separating water vapor from the carbon dioxide. The CO_2 is then condensed for transportation.

Figure 3:



Carbon capture and storage – oxyfuel-combustion (Oxyf) process [1,4,6]

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For all methods, CO_2 is typically compressed to supercritical state (~10 MPa) for transportation and storage [7]. For transportation, pipes are continuously being built and the transport of compressed CO_2 has relatively low costs. Over 5,000 miles of CO_2 pipelines exist in the United States at the end of 2018 [5].

Storage can typically be done in geologic formations, such as depleted oil and gas fields, saline formations, unmineable coal seams [1]. There is also the possibility of storage in the deep ocean, injecting liquid carbon dioxide at intermediate (1000-3000 m) or depths greater than 3000 m to form a "CO₂ lake" at the bottom of the ocean [2]. Storage is plentiful, as it is estimated there is 1120-3400 billion tonnes of CO₂ possible to be sequestered [1]. Injecting CO₂ into oil and gas fields is already a practiced technology, but the environmental impacts (leaks, slow migration and accumulation) are still being studied for many geologic storage systems [2].

CCS Method Process Flows

Energy and material process flow diagrams were created for the three different carbon capture systems, based on primary power plant type and size. For all plants, the analysis is based on numerical estimates, as there have been no plants built with CCS systems in the original design (not retrofitted). Retrofitted systems typically capture less carbon and require more energy input, as they are not the most efficiently designed.

All systems require some amount of electrical power (for air separation units and carbon capture methods). This can be taken directly from the power plant's electrical output. However, this lowers the plant's efficiency and increases total electricity and fuel costs to keep it operating at the same power level. Alternatively, natural gas-fired combined cycle can be installed to provide electricity for the carbon capture systems, as is done in the case of the Petra Nova plant in Thompsons, TX [19].

Figure 4 [4,6,8] describes a theoretical new (not retrofitted) post-combustion capture system on a pulverized coal power plant, nominally operating at 592 MWe. This is an MEA (amine-based capture system) at 2020 technology levels [6]. Water inputs and outputs were not given in this scenario, so a 685 MWe pulverized coal plant with the same amine-based capture system was downsized to the smaller plant, assuming similar fractional water inputs and outputs [8]. Note that the power required for the CCS system alone is 172 MWe.



Figure 4:

PCC energy and material flow process diagram in a new pulverized coal power plant (built with PCC instead of retrofitting) [4,6,8]

Next is the pre-combustion/IGCC input and output diagram, as shown in Figure 5 [4,9]. The plant is an IGCC cogeneration plant with CCS, noting that this is a conceptual plant as pre combustion capture is still a developing technology. The model is based on a 250 MWe IGCC without CCS [9]. Note that water and

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chemical outputs were not given in this study. Also note that the power requirement for carbon capture is significantly lower than that on the PCC, even though the plant size is only half.

Figure 5:



Input and output diagram for IGCC cycle in a model IGCC cogeneration plant [4,9]

Last is the Oxyf technology system, shown in Figure 6 [4,10]. The inputs and outputs are based on a supercritical hard coal power plant (operating with steam at 600 °C and 285 bar), and assuming oxyfuel technologies with a cryogenic air separation unit and 95% oxygen [10]. This plant outputs 440.9 MWe and would require 93 MWe of power to operate the carbon capture system.

Figure 6:



Material and energy flow for a conceptual Oxyf combustion CCS system in a supercritical hard coal power plant, assuming 95% pure oxygen flow [4,10]

A comparison between carbon capture and storage methods, normalized to power output was also done and is shown in Table 1. Note that these describe different power plant operations (post-combustion capture is used on

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a pulverized coal power plant, oxyfuel on a supercritical hard coal plant, and pre-combustion on an integrated gasification cogeneration power plant). Also note that it was difficult to find all inputs and outputs for IGCC systems as they are mostly based on numerical estimates as opposed to experimental values, but it can be assumed that water and waste are similar to PCC and Oxyf technologies. The additional power requirement for oxyfuel is lowest (as a percentage of total input power), likely due to the simple capture technology (with no chemical or physical solvents required). The energy required for these capture systems may come from a separate power generating facility near the plant, such as natural gas. This helps decrease the additional amount of carbon dioxide that must be captured to run the CCS system.

Inputs (t/MWd)	PCC	IGCC	Oxyf
Coal	10.34	9.04	9.49
Water	84.26	36.99	70.13
MEA (PCC) /MDEA (Oxyf)	0.027	0.001	
NH ₃	0.015		0.005
NaOH	0.002		
Limestone Additional Energy Required for Same Power	0.58		0.17
	29%	25%	21%
Outputs (t/MWd)			
CO ₂ Emission	2.71	2.26	2.23
CO ₂ Capture	24.41	23.78	21.29
CO ₂ Capture Percentage (%)	90%	91%	91%
Water	24.91		23.28
Waste (ash, gypsum, heavy metals)	1.07		1.63
CH ₄	0.075		
NH ₃	0.004		
SO ₂	0.006		0.00001
NO _x	0.019		0.001
СО			0.002

Table 1. Inputs and outputs for average carbon capture and storage systems (PCC, IGCC, and Oxyf), measured in t/MWd[4,6,8,9,10]

The results for CO_2 capture and emission rates from Table 1 are plotted in Figure 7. It is clear from this graph that all methods capture and release similar amounts of carbon dioxide. While Oxyf captures the least amount of carbon dioxide, it also emits the least amount. It can also be shown from Table 1 that much of the basic inputs and outputs (coal, water, and CO_2 emission and capture) are all relatively similar between the three CCS systems. PCC requires the most energy, likely due to the chemical separation of low partial pressure CO_2 using amine based systems. Oxyf releases the smallest amount of undesireable chemicals besides carbon dioxide (SO₂ and NO_x). All in all, the systems are relatively similar, with the main difference being the lower technolgical readiness levels for IGCC and Oxyf, especially compared to PCC.

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[Pinsky * et al., 8(7): July, 2019] ICTM Value: 3.00 Figure 7:



CCS CO₂ Capture and Emissions in t/MWd

CO₂ capture and emission comparisons for various CCS systems.

Cost Comparison

CO2 capture represents the highest cost to CCS, where the majority of the cost increase is due to capital investments in a capture system [11]. Table 2 shows a cost analysis for the three CCS systems from 2011, for first of a kind plants designed with these systems [11]. This shows Oxyf having the lowest levelized cost of production, but not the lowest increase from a system without CCS. The levelized cost is based on a levelization period of 30 years with constant 2010 US currency.

	PCC	IGCC	Oxyf
Levelized Cost of Production with CCS (\$US ₂₀₁₀ /MWh)	120-131	125	114-123
% Increase in Levelized Cost than without CCS	61-76%	37%	53-65%
Cost of CO ₂ Avoided (\$US ₂₀₁₀ /t CO ₂)	62-81	67	47-59
Cost of CO ₂ Captured (\$US ₂₀₁₀ /t CO ₂)	53-55	39	42-47

Table 2. Cost analysis of various CCS systems (PCC, IGCC, and Oxyf) [11]

The study also indicates the cost of carbon dioxide as a breakpoint. For Oxyf CO₂, the credit value breakpoint is approximately \$US₂₀₁₀55/t of CO₂ (in 2010) [11]. For an IGCC system with supercritical pulverized coal technology was at \$70/t of CO₂, and for natural gas at \$112/t of CO₂, due to the lower emission intensity of natural gas and higher efficiencies relative to coal [11]. For PCC in a supercritical pulverized coal plant, the cost is around \$80/t of CO₂ [11]. In the same study, transportation was estimated to be between \$1-2/t of CO₂, and storage was anywhere from \$6-13/t of CO₂. Note that the cost of transportation and storage is relatively small compared to the cost of capture. Also note that these costs are decreasing over time with improvements in technology. Other studies have shown similar values for the costs of CO₂ necessary for CCS technologies to be adopted into the power production industry [2].

There are also federal policies developing in the United States aimed at giving tax credits to the carbon capture industry [5]. The 45Q tax credit, for example, recently passed providing \$35 to \$50 per metric ton of CO₂ emissions reduced using CCS (and beneficially used or stored in geologic formations) for up to 12 years after being in service [5]. The minimum threshold to claim these benefits was also lowered to 100,000 tons of CO₂ captured per year [5]. For a nominal PCC plant, this would save around \$185 million per year. If a retrofit PCC system costs around \$2 billion to install for a nominal plant, this would take just under 11 years to pay off from this tax credit [12].

3. FUTURE CARBON REDUCCTION METHODS

In addition to CCS processes that can be retrofitted to power plants, methods are being developed to capture carbon from alternative sources. The carbon that is captured may be stored like previously mentioned CCS

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systems, or it may be utilized to form new products. Direct air capture is a developing technology that would be able to capture carbon dioxide from the air. An important utilization technology comes from the company LanzaTech, which created a microbe that ferments carbon syngas to produce ethanol. There are a multitude of other carbon utilization systems from infancy to commercial maturity [13]. Other carbon utilization schemes include mineral carbonation processes to produce construction materials and chemical and biological conversion to produce chemicals, fuels, or polymers [13].

Direct Air Capture

Direct air capture uses solvents to capture CO_2 directly from the atmosphere. Instead of attempting to address large scale point sources of carbon that the aforementioned CCS technologies use, it could be available everywhere. Direct air capture has the advantage of being a truly carbon negative technology, as it does not require a carbon source [14]. The process for this system is virtually the same as the PCC carbon capture system using amine solvents. The anime solvents have been proven to work for concentrations of CO_2 as low as atmospheric 390 ppm levels, though with somewhat reduced efficiencies [14].

Though the capture technology has been proven to work at atmospheric concentrations, it is not economically viable currently. Although direct air capture has the potential to work on the large-scale levels, it could cost 600 or more per metric ton of CO₂ [15]. However, there are significant research efforts underway to improve direct air capture efficiencies and it is a promising technology. Direct air capture may be economically effective as a carbon reduction strategy when large centralized sources of carbon (i.e. power plants) are eliminated [15].Even still, the direct air capture venture is underway as of 2019. Oxy Low Carbon Ventures, LLC, and Carbon Engineering, Ltd have recently announced plans to evaluate a direct air capture facility capable of capturing up to 500 kt CO₂ per year (the largest in the world) [20]. The facility will be designed to be eligible to the previously discussed 45Q tax credits to reduce costs [20].

Alternative Fuel Production with Syngas Fermentation

Biological routes of carbon capture are currently being investigated and range from photosynthetic, such as algae cultivation [21], to non-photosynthetic, for example acetogenic micoorganisms [22,23]. An example of a technology that is currently being piloted at scale is based on a microbial gas fermentation. The company LanzaTech has created proprietary microbes that can convert syngas into several chemicals or fuel, specializing in ethanol [16]. By converting combinations of hydrogen and carbon monoxide, it is possible to produce low CO_2 emission fuels. Many industrial processes, such as steel manufacturing, produce CO containing gases as a byproduct [16]. CO leaking into the atmosphere contributes to greenhouse gas emissions, so reducing this pollutant could help minimize the environmental damages from this process.

The LanzaTech process is shown in Figure 8. The syngas is compressed to optimal fermentation conditions, at a pH of 4-6, temperature of 35-42 °C, and pressure of 0-5 bar [17]. Then, the gas is fed to the reactor where it is fermented using catalysts and proprietary microbes. The products are then recovered, processed, and separated to obtain the desired compounds, such as ethanol.

Figure 8:



LanzaTech syngas fermentation process [16]

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The feed gas for the LanzaTech process can be created in a multitude of ways. It can be produced from the gasification of some product, whether coal, petroleum, natural gas, biomass, or solid waste. It is also produced from certain industrial waste streams, such as from steel mills or chemical plants [18]. Note that the IGCC process includes a gasification step that produces syngas. Also note that the syngas feedstock for gas fermentation can be a variety of CO to H_2 ratios. In other conversion devices, such as thermochemical conversion to ethanol, impurities must be cleaned and the ratio of gas feed must be precise [17].

One of the main objectives of gas fermentation is to combine it with a clean source of feed gas. This is typically from biomass conversion. Gas fermentation from biomass would produce around 1.46 kg CO_2 per kg of feedstock input, at a feedstock flow rate of 2000 metric tons per day, producing 672 tons per day [17]. The emissions from the ethanol are significantly lower than what is given off in other fuel combustions (such as the coal in a power plant). However, a disadvantage for biomass conversion resides in scalability. Currently, the largest biomass gasifiers are restricted to 1000-2000 tons per day of feed due to biomass gasifier technology and collection limits [17]. The LanzaTech technology has demonstrated scalability for commercial development to attached to gaseous waste stream industrial processes, and biomass gasification is still under development.

4. CONCLUSION

Carbon capture and storage is a potential way to greatly limit the release of CO_2 in the air in industries that have significant contributions to emissions. Limiting emissions is necessary to mitigate the acceleration of global climate change. Though zero emission renewables are preferable to the fossil fuel industry, the current and near future society relies on fossil fuels for power production. CCS systems can be applied in three main ways: pre/post-combustion and oxy-fuel.

Currently, PCC is the most developed technology, actively being retrofitted to power plants, but it does not fare best in terms of energy requirements or levelized cost. Oxyfuel technology has the highest potential, as it does not require energy intensive capture technologies, and has the lowest levelized cost of production. However, oxyfuel technology is still in early technological development.

All CCS systems provide more benefits than downsides. Though all require a non-insignificant amount of electricity for operation, and a significant amount of water, they also reduce CO_2 emissions by over 90%. However, CCS is a potentially very beneficial technology for the purpose of emission reduction. CCS already has over 25 million metric tons of capacity in the United States as of 2018 and is continuously developing as a technology [5].

Finally, alternative methods are under development for further carbon capture and utilization. Direct air capture has been proven effective but currently are faced with high costs when compared to the relatively inexpensive costof placing carbon capture at large sources of CO_2 , such as a power plant. Biological carbon capture approaches are also being investigated that utilize either waste gas streams (such from a coal-fired power plan or other industrial process) or gasification of a biomass source (such as woody biomass or municipal solid waste). Current biomass gasification technologies limit production using this method to small scales.Significant advances in these alternative approaches to carbon capture are likely, which will increase their utilization at industrial-scales.

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