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Assessment of the Current State of Carbon Capture for Climate Change Mitigation

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The world will not be destroyed by those who do evil, but by those who watch them without doing anything.

Albert Einstein

I would like to express my sincere gratitude to my tutor, Dra. Carme Sans Mazón, for their invaluable guidance, support, and expertise throughout the course of this research. Their insightful feedback, patience, and encouragement have been instrumental in shaping the direction and quality of this paper.

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CONTENTS

SUMMARY

Carbon capture technologies are considered an important strategy to mitigate climate change by removing carbon dioxide from effluents of industrial sources, particularly power plants. This project compares three prominent carbon capture technologies in terms of their efficiency in removing carbon dioxide, their economic costs, and their environmental impacts. The findings showed that each technology possesses advantages and disadvantages.

Post-combustion carbon capture using an amine solvent was shown to be most developed technology, however a literature review revealed many problems regarding high energy consumption, high corrosivity and high toxicity (particularly due to degradation products).

Pre-combustion carbon capture was demonstrated to be a potentially more efficient carbon capture method due to gasification which produces a synthesis gas with high pressure and high CO_2 concentration, from which the CO_2 is captured. However, the gasification process adds complexity and has a high energy penalty, which reduces the viability of the process.

Oxy-fuel combustion was revealed to be a promising technology which vastly increases the CO_2 in the flue gas by combusting the fuel with oxygen of high purity, from an air separation unit (ASU). The requirements of oxygen for the ASU present a unique challenge for oxy-fuel combustion carbon capture.

Furthermore, the sequestration of carbon dioxide presents many challenges including leakage and a high storage requirement to achieve climate change mitigation goals. Additionally, enhanced oil recovery was shown to be an alternative to sequestration, which offsets the benefits of carbon capture for climate change mitigation.

Life Cycle Assessments revealed that post-combustion capture with an amine solvent has negative environmental effects that greatly offset its benefit. Techno-Economic Analyses demonstrated that all carbon capture technologies result in a considerable efficiency loss.

In conclusion, this project highlights the need for further development of carbon capture to improve their ability to mitigate climate change. Particularly, research is needed to reduce the energy demands for gasification for pre-combustion capture and the ASU for oxy-fuel capture. More imperatively, more research is required to reduce leakage from sequestration sites of captured carbon dioxide. Moreover, this project highlights the importance of policy for carbon capture to be viable for climate change mitigation, particularly the redirection from the use of enhanced oil recovery to dedicated geological storage of captured carbon dioxide. At its current state, carbon capture was not seen to be viable for mitigating climate change, however it was demonstrated to be a promising technology.

Keywords: climate change, carbon capture, post-combustion capture, pre-combustion capture, oxy-fuel combustion capture, carbon dioxide sequestration, enhanced oil recovery, mitigation strategies.

RESUMEN

Las tecnologías de captura de carbono se consideran una estrategia importante para mitigar el cambio climático al eliminar el dióxido de carbono de los efluentes de fuentes industriales, especialmente las plantas de energía. En este proyecto, se comparan tres tecnologías destacadas de captura de carbono en términos de su eficiencia para eliminar el dióxido de carbono, sus costos económicos y sus impactos ambientales. Los hallazgos mostraron que cada tecnología tiene ventajas y desventajas.

La captura de carbono postcombustión (post-combustion carbon capture en inglés) mediante un solvente de amina se demostró como la tecnología más desarrollada; sin embargo, una revisión bibliográfica reveló numerosos problemas relacionados con el alto consumo de energía, la corrosividad y la toxicidad (particularmente debido a los productos de degradación).

La captura de carbono precombustión (pre-combustion carbon capture en inglés) se demostró como un método potencialmente más eficiente debido a la gasificación, que produce un gas de síntesis con alta presión y alta concentración de CO_2 del cual se captura el CO_2. Sin embargo, el proceso de gasificación añade complejidad, lo que reduce la viabilidad del proceso.

La combustión con oxígeno puro (oxy-fuel combustion carbon capture en ingles) se reveló como una tecnología prometedora que aumenta considerablemente el CO_2 en los gases de combustión al quemar el combustible con oxígeno de alta pureza de una unidad de separación de aire (ASU). Los requisitos de la ASU plantean un desafío único para la captura de carbono mediante la combustión con oxígeno puro.

Además, la captura y almacenamiento de dióxido de carbono presenta muchos desafíos, como las fugas y la alta capacidad de almacenamiento necesaria para lograr los objetivos de mitigación del cambio climático. Además, se demostró que la recuperación mejorada de petróleo

es una alternativa a la captura y almacenamiento de carbono, lo cual contrarresta los beneficios de la captura de carbono para la mitigación del cambio climático.

Los análisis del ciclo de vida revelaron que la captura de carbono posterior a la combustión con un solvente de amina tiene un impacto ambiental negativo que compensa en gran medida sus beneficios. Los análisis tecno-económicos demostraron que todas las tecnologías de captura de carbono resultan en una pérdida considerable de eficiencia.

En conclusión, este proyecto destaca la necesidad de un mayor desarrollo de la captura de carbono para mejorar su capacidad de mitigar el cambio climático. Especialmente, se requiere investigación para reducir las demandas de energía de la gasificación en la captura previa a la combustión y de la ASU en la captura mediante la combustión con oxígeno puro. Además, se necesita más investigación para reducir las fugas en los sitios de almacenamiento del dióxido de carbono capturado. Además, este proyecto resalta la importancia de las políticas para que la captura de carbono sea viable para la mitigación del cambio climático, especialmente en la transición del uso de la recuperación mejorada de petróleo hacia el almacenamiento geológico del dióxido de carbono capturado. En su estado actual, no se considera que la captura de carbono sea viable para mitigar el cambio climático, sin embargo, se ha demostrado que es una tecnología prometedora.

Palabras clave: cambio climático, captura de carbono, captura posterior a la combustión, captura previa a la combustión, captura mediante la combustión con oxígeno puro, secuestro de dióxido de carbono, recuperación mejorada de petróleo, estrategias de mitigación.

SUSTAINABLE DEVELOPMENT GOALS

Este trabajo está estrechamente vinculado al ámbito del medio ambiente, específicamente al Objetivo 13: Acción por el Clima. El proyecto fue seleccionado en respuesta a las consecuencias actuales del cambio climático, incluyendo la sequía que está experimentando Catalunya. El objetivo de este proyecto es investigar tecnologías para reducir las emisiones generadas por los procesos industriales. Además, las consecuencias del cambio climático también afectan significativamente a los ecosistemas terrestres y marinos, por lo tanto, este proyecto también se relaciona con los Objetivos 14 y 15 (Vida Submarina y Vida de Ecosistemas Terrestres).

En este proyecto también se evalúan los costes y la eficiencia de las tecnologías de captura de carbono, investigando si estas tecnologías pueden proporcionar energía asequible. También se investigan los efectos que estas tecnologías producen en el entorno. Por lo tanto, se vincula al Objetivo 7 (Energía asequible y no contaminante).

Además, este proyecto fomenta la innovación tecnológica, ya que es fundamental para el éxito de la tecnologías de captura de carbono. Esta innovación podría generar más empleo y crecimiento económico. En consecuencia, se relaciona con el Objetivo 9 (Industria, Innovación e Infraestructura) y el Objetivo 8 (Trabajo decente y crecimiento económico).

Sin embargo, es importante destacar que este proyecto no aborda en gran medida el ámbito de las personas (People). El proyecto demuestra que las tecnologías de captura de carbono podrían resultar en un aumento del costo de la energía, lo que probablemente tendría un impacto en la sociedad. Este efecto sería más pronunciado en los países en desarrollo, que probablemente no podrían permitirse gastar más en energía. No obstante, esto está fuera del alcance de este proyecto, cuyo objetivo principal es investigar cómo es la forma más eficiente de reducir las emisiones.

Tampoco se discute el ámbito de la paz (Peace). Aunque existen muchos efectos sociales del cambio climático que entran en estos ámbitos, debido a que los desastres medioambientales pueden provocar disturbios civiles, no se hace referencia a estos efectos dentro de este proyecto.

Otros proyectos podrían investigar cómo la tecnología de captura de carbono podría tener efectos en los ámbitos de las personas (People) y la paz (Peace).

1. INTRODUCTION

This project is focused on carbon capture technologies, which remove carbon dioxide through various methods from emitting sources in power plants and other industrial sources. The captured carbon dioxide can be stored or used in other industrial processes, which reduces the carbon dioxide emitted.

1.1. THE CURRENT CLIMATE CHANGE CRISIS

The context of the project is the urgent need to address climate change in accordance with the Paris Agreement, a global framework aimed at limiting global warming to the range of 1.5°C-2°C, which requires capping atmospheric CO_2 levels to around 450ppm. (Bui et al., 2018; European Commission, 2015). UN backed research indicates that a 45% reduction in global emissions is required by 2030, yet current projections suggest that emissions will increase by 14% by that time (United Nations Framework Convention on Climate Change, 2022). The UN has stated that achieving carbon neutrality before 2050 is necessary to prevent further global temperature rise, which is currently projected to reach 3.2°C by 2100 if current policies continue (Intergovernmental Panel on Climate Change, 2023).

A consequence of global warming is rising sea levels. NASA has measured changes in the Global Mean Sea Level using satellite altimeters for the past 27 years (1993-2009). Currently, global sea levels are rising 3.4 millimetres and in those 27 years, the total sea level has risen by 88.9mm**.** (NASA, n.d.)

Moreover, the vulnerability of both humans and ecosystems is mutually dependent, as the rise in occurrences of extreme weather and climate events has led to acute food insecurity and reduced water availability for millions of individuals. Around 3.3-3.6 billion people live in contexts

that are highly vulnerable to climate change, particularly communities in Africa, Asia, Central and South America. Regions that were highly vulnerable to floods, droughts, and storms experienced a 15 times higher human mortality rate between 2010 and 2020 compared to regions with very low vulnerability (Intergovernmental Panel on Climate Change, 2023).

1.2. THE PROPOSED SOLUTIONS TO MITIGATE CLIMATE CHANGE

1.2.1. Current Technology to Mitigate Climate Change

The United Nations (2023) states that "fossil fuels, including coal, oil and gas, account for over 75% of global greenhouse gas emissions and nearly 90% of all carbon dioxide emissions.". There are many technologies that can replace fossil fuel combustion to reduce greenhouse gas emissions and thus, mitigate climate change.

These include renewable energies which don't require fossil fuels and don't directly emit greenhouse gases, such as wind, solar and hydropower, which is expected to comprise 40% of all energy production in 2027 (based on an IEA analysis with wind and solar comprising over 90% of that renewable energy growth in the next five years) (International Energy Agency, 2022b).

Another alternative is nuclear, which doesn't directly emit greenhouse gases despite having high levels of indirect emissions because of mining and construction (International Energy Agency, 2022b). Globally, around 10% of electricity production comes from nuclear with some countries having higher usage such as France with 70% of the electricity being produced from nuclear (Ritchie, Roser and Rosado, 2022).

Energy efficiency measures are also highly effective in reducing greenhouse emissions as they are based on readily available cost-effective technologies. These measures cover a broad range including the transfer from internal combustion engines to electric vehicles, the adoption of smart meters, appliances and sensors and the replacement of old inefficient equipment with new compliant (with energy saving programs) equipment (International Energy Agency, 2022b).

1.2.2. The Necessity for Carbon Capture Technologies

Carbon capture technologies exist as another option for reducing carbon dioxide emissions from power generation and other industrial processes.

An analysis by the International Energy Agency (2020) showed that achieving net-zero goals will be virtually impossible without widespread adoption of carbon capture technologies. As such, the development and implementation of carbon capture is considered a crucial step in achieving global climate goals and mitigating the impact of climate change. To stay on the path to meet the 2 °C targets, at least 90 gigatonnes of CO_2 emissions must be reduced by countries belonging to the United Nations Economic Commission for Europe (UNECE) by 2050 (United Nations Economic Commission for Europe, 2021).
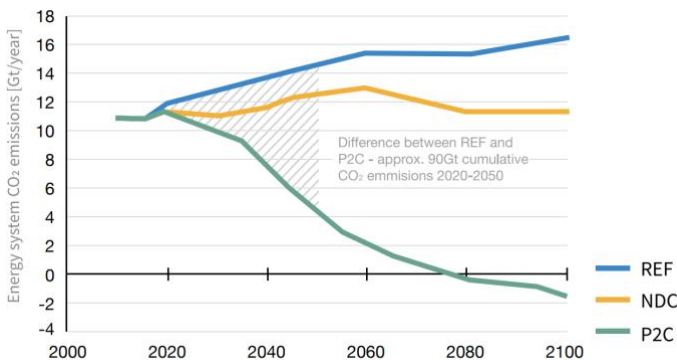

Figure 1: CO2 emissions in the UNECE region by policy scenario for the energy sector. REF, NDC, P2C represent three different forecasts for CO2 emissions *(image retrieved from United Nations Economic Commission for Europe, 2021)*

The above graph shows the CO_2 emissions in the UNECE area by various policy scenarios: REF, the forecast of CO_2 emissions based on maintaining economic growth; NDC, a scenario in which the constraints imposed by Nationally Determined Contributions under the Paris Agreement up to 2030 are enforced and maintained indefinitely; and P2C; which constrains emissions to limit global warming to less than 2 °C.

Currently, due to inaction in reducing carbon dioxide emissions, many models now include negative emissions, which is the removal of carbon dioxide from the atmosphere. This is shown in the P2C scenario in Figure 1, which incorporates negative emissions in the future in its model. The necessity of negative emissions in these models indicates that carbon capture alone may not be sufficient to mitigate global warming to 2 °C.

A vast amount of CO_2 is emitted in the energy production sectors, specifically for electricity and heat. Therefore, carbon capture technologies are primarily focused on reducing emissions from power generation, which makes it the main focus of this project.

In conclusion, the effects of climate change are already being felt and the Paris Agreement and the United Nations' call for carbon neutrality before 2050 highlight the seriousness of the situation. Carbon capture technologies are seen as a crucial step in mitigating the impact of climate change, but their effectiveness must be assessed at an industrial scale.

2. OBJECTIVES

The general objective of this project is to evaluate the feasibility of various carbon capture technologies in achieving climate change targets. In particular, the project has the following specific objectives:

- A comprehensive review of the current state of carbon capture technology, exploring all the options currently available. Despite this, more research is to be dedicated to technologies that have undergone more research and development.
- Comparison of the main carbon capture technologies based on findings from life cycle assessments and techno-economic analyses.
- Investigate the viability of carbon capture storage and how this affects the net emissions of the carbon capture process.
- Critically assess the effectiveness of carbon capture as a strategy for mitigating climate change, taking into account economic, social, and environmental considerations.

3. THE CARBON CAPTURE PROCESS

In this section, the current state of carbon capture technology shall be assessed regarding their effectiveness at reducing emissions. Carbon capture is often synonymous with carbon capture and storage (CCS), which encompasses the capture of carbon dioxide, as well as the transport and storage of the CO_2. The following Figure 2 shows a brief view of the available carbon capture technologies as well as the storage and utilisation options available for captured CO_2. The primary methods will be covered in this project.
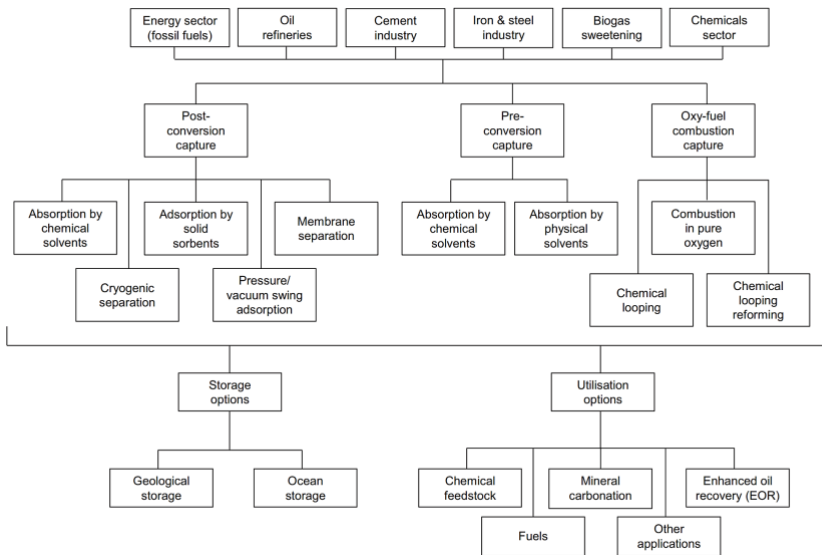

Figure 2: Different carbon capture, storage, and utilisation options *(image retrieved from Cuéllar-Franca and Azapagic, 2015)*

There are three main configurations of carbon capture technologies: post-combustion capture technology, pre-combustion capture technology and oxy-fuel combustion capture technology. The carbon capture process itself is considerably more expensive than the transport and storage of the captured CO_2, incurring about 75% of the cost (Yadav and Mondal, 2022).

As a quick summary, post-combustion involves the capture of carbon dioxide from the flue gas produced from combustion whereas pre-combustion captures carbon dioxide before combustion is undergone. Oxy-fuel combustion involves combustion of the fuel at high oxygen content so the carbon dioxide can be easily separated.

The Global CCS Institute reported there were 196 CCS facilities in various stages of development at 2022 (Steyn et al., 2022). CCS refers to carbon capture and storage and, in this context, is interchangeable with the term carbon capture used in this project. Figure 3 shows the number of carbon capture facilities in each stage of development.
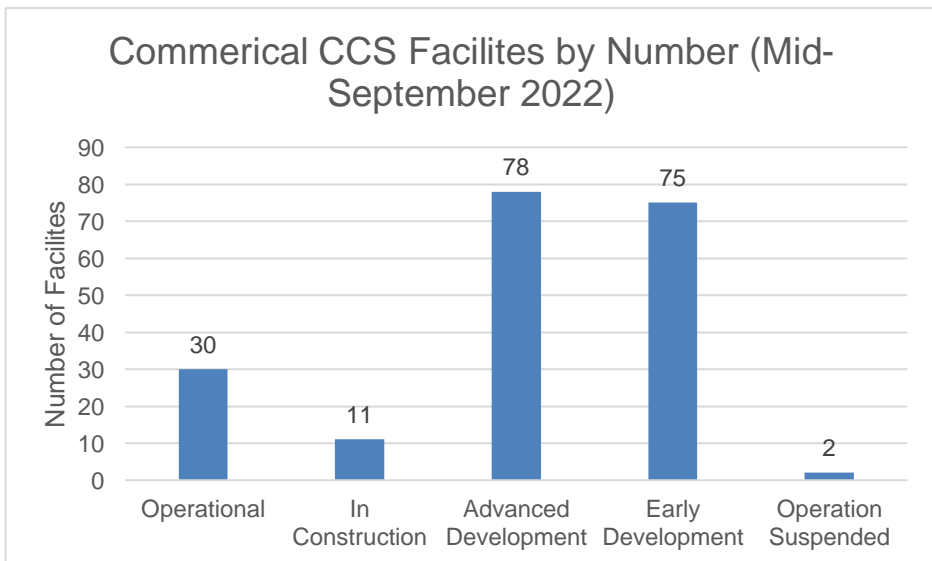

Figure 3: Number of CCS facilities categorised by stage of development as of September 2022
(using data from Steyn et al., 2022)

3.1. POST-COMBUSTION CARBON CAPTURE

Post-combustion carbon capture has many forms, which primarily depend on the solvent or material used to extract the carbon dioxide to be pressurised and transported: chemical solvents, solid solvents, and membranes.

The major advantage that all forms share is that the pollution control focuses on the final stage of the industrial process, and thus, possesses "end of pipe" characteristics that make these types of facilities easily retrofittable to existing units (Wang et al., 2017; Figueroa et al., 2008). However, since little modification is done before the fuel is combusted, the CO_2 in the resulting gas exists at dilute concentration, which depends on the fuel being combusted. Additionally, there are impurities of SO_2 and NO_2 in the flue gas.

Consequently, there is a low thermodynamic driving force and thus, a large volume must be treated, and the captured CO_2 must be compressed from atmospheric to pipeline pressure. (Figueroa et al., 2008; Scheffknecht et al., 2011).

3.1.1. Absorption by Chemical Solvents

Absorption can be separated into two categories: chemical and physical. In chemical absorption, a reaction takes place with CO_2 and then the solvent is regenerated by heating to desorb the CO_2. In physical absorption, the solvent does not react with CO_2 and the driving force for CO_2 separation is the solubility of CO_2 in the solvent. Chemical absorption is more favourable for capturing CO_2 at relatively low pressure and thus, is used predominately for post-combustion carbon capture where the flue gas is at low pressure (Babar et al., 2019; Sifat and Haseli, 2019). As the flue gas in post-combustion has low CO_2 concentration and is at low pressure, physical absorption is more relevant to pre-combustion carbon capture. The reasons for this will be discussed in Section 3.2.1.
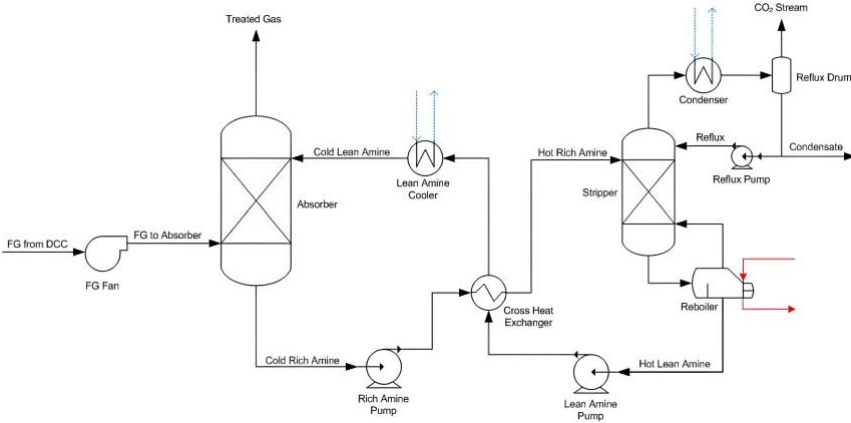
Figure 4: Basic flow sheet for an amine-based carbon capture process *(image retrieved from Agbonghae et al., 2014)*

The amine-based carbon capture process follows a flow sheet depicted in Figure 4 (Agbonghae et al., 2014). The process involves counter-current contact of the flue gas, cooled in a direct contact cooler (DCC) unit, with a chemical solvent (typically an amine solvent) which absorbs the carbon dioxide.

The flue gas enters at the bottom of the absorber, while the lean amine solution enters the top. The rich solvent from the bottom of the absorber is then sent to the stripper; as it is sent, the solvent absorbs heat from the lean solvent exiting the bottom of the stripper in a cross-heat exchanger. In the stripper, the CO_2 is stripped from the rich solvent by the upward flowing steam generated in the reboiler. The vapour stream (composed of CO_2, steam and small amounts of the amine) at the top of the stripper is partially condensed in a condenser. The condensed liquid is then returned to the top of the stripper as reflux, while the uncondensed stream, mainly carbon dioxide, is compressed, transported, and sequestered (Agbonghae et al., 2014).

The regeneration of the solvent accounts for the most energy consumption and represents a crucial focus area in carbon capture solvent research.

Typically, this solvent is an alkanolamine solvent. This is because amines react rapidly and selectively with CO_2 and are relatively inexpensive (Songolzadeh et al., 2014). Mono-ethanol amine (MEA), di-glycol amine (DGA), di-ethanol amine (DEA) and methyl di-ethanol amine (MDEA) are commonly used, of which MEA is the most utilised due to its capability of achieving

high level of CO_2 capture resulting from its fast kinetics and strong chemical reaction with CO_2 (Seidi et al., 2019; Polasek and Bullin, 2006). Currently, MEA is the most researched and utilised solvent for carbon capture and thus is the focus for this study of amines (Dutcher, Fan and Russell, 2015).

The amines can be classified into three types: primary, secondary and tertiary amines that correspond to the number of hydrogen atoms that are linked covalently to the nitrogen atom: two, one and zero, respectively. The chemical reactivity of the amines is in descending order: primary ($-NH_2$), secondary ($-NH$) and tertiary ($-N$) (Yamada, 2021). MEA and DGA are primary amines, DEA is a secondary amine and MDEA is a tertiary amine.
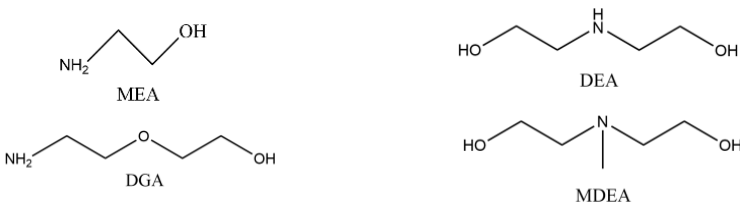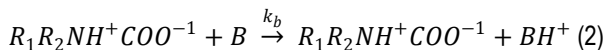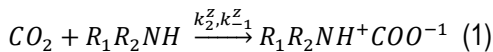

Figure 5: Chemical structures of MEA, DEA, DGA and MDEA (generated with *ChemDraw*)

The reaction mechanism of CO_2 absorption by an aqueous amine solution, first proposed by Michael Caplow in 1968, includes the zwitterion mechanism, the termolecular mechanism and the base-catalysed hydration mechanism (Chen et al., 2023; Hu et al., 2022)

The process begins with the creation of zwitterionic intermediates as the first step and in the subsequent step, these zwitterionic intermediates undergo deprotonation, resulting in the formation of carbamates. The base (B) can be R_1R_2NH (primary amine and secondary amine), $OH-$ or H_2O (Chen et al., 2023)

$$CO_2 + R_1R_2NH \xrightarrow{k_2^z, k_{-1}^z} R_1R_2NH^+COO^{-1} \quad (1)$$

$$R_1R_2NH^+COO^{-1} + B \xrightarrow{k_b} R_1R_2NH^+COO^{-1} + BH^+ \quad (2)$$

In the case of MEA, a breakdown of the C-N bond of MEA carbamate occurs, forming a Zwitterion, which can decompose into MEA, water, and CO_2.

$$MEA - COO^{-1} + H_3O^+ \leftrightarrow MEA + H_2O + CO_2 \quad (3)$$
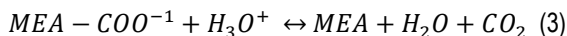
The deprotonation of MEA-H$^+$ is another very important reaction.

$$MEA{-}H^+ + H_2O \leftrightarrow MEA + H_3O^+ \quad (4)$$
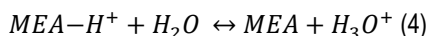
The processes described in reactions 3 and 4 are very endothermic and this, coupled with the difficulty of proton transfer in 4 results in an elevated energy cost for the MEA solution regeneration process (Hu et al., 2022).

The following Table 1 summarises the main advantages and disadvantages of the amine solvents, obtained by bibliographic review.

Table 1: Summary of the main differences between the most used amine solvents

Amine	Advantage	Disadvantage	Reference
MEA (primary)	■ Most studied and developed amine. ■ Exhibits fast kinetics and strong chemical reaction with CO_2.	■ Degradation products are corrosive and toxic. Easily degrades with impurities of SO_2 and O_2. ■ Reacts with oxidizing agents producing corrosive and soluble products. ■ High heat of reaction with CO_2. ■ High regeneration energy requirement.	(Dutcher, Fan and Russell, 2015), (Luis, 2016), (Polasek and Bullin, 2006), (Supap et al.,2009), (Uyanga and Idem, 2007)
DEA (primary)	■ Degradation products are less corrosive than those of MEA.	■ Reduced affinity for CO_2 (and H_2S). ■ Degradation products are less corrosive than those of MEA.	(Newpoint Gas, LLC, 2016), (Polasek and Bullin, 2006), (Xue et al., 2016)
DGA (secondary)	■ Higher concentrations of DGA result in lower circulation rates and lower freezing points.	■ High heat of reaction for CO2 (and H2S).	(Polasek and Bullin, 2006)
MDEA (tertiary)	■ Readily selective toward H_2S in the presence of CO_2. ■ Lower vapour pressure.	■ Lower reactivity as a tertiary amine.	(Polasek and Bullin, 2006), (Yamada, 2021)

■ High degradation resistance
(thermal and oxidative).

■ Lower corrosivity.

The literature review reveals that MDEA has distinct advantages over the other alkanolamines but lacks reactivity. The high reactivity of MEA makes it the most utilized solvent for carbon capture. Mixed amines (including MDEA/MEA) are a promising field as they retain the reactivity of primary amines but offer lower regeneration rates (Idem et al., 2006).

A study conducted by Gunasekaran, Veawab and Aroonwilas (2017) showed that the corrosivity order of carbon steel (specifically CS108) in single amine systems saturated with CO_2 at 80 °C decreases in the following order: MEA>DEA>MDEA (DGA was not included in this study). Using 5.0 kmol/m^3 MEA at 0.20 mol CO_2/mol amine loading, the study evaluated the corrosion at three different temperatures and showed that the corrosion rate increases with temperature, following Arrhenius behaviour. This direct relationship is shown in the following Figure 6.
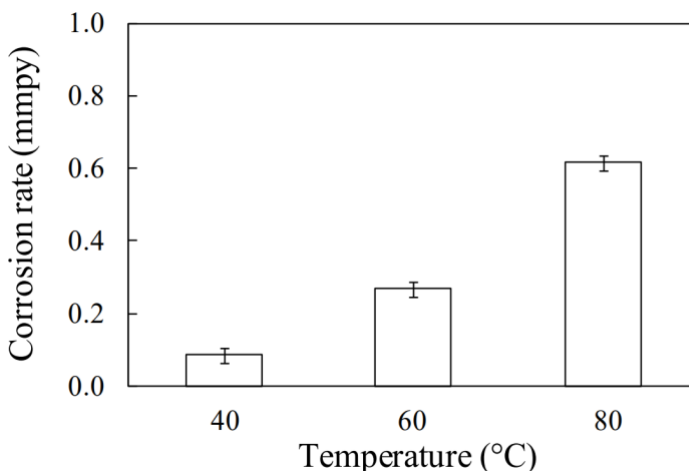

Figure 6: Effect of temperature on corrosion rate of CS108 in 5.0 kmol/m3 MEA at 0.20 mol CO_2/mol amine loading (image retrieved from Gunasekaran, Veawab and Aroonwilas, 2017).

The corrosivity of the amines has different effects on different parts of the plant. This was shown in a pilot plant study undergone by Kittel et al. (2009), which used probes in various positions of the CO_2 capture plant and tested the corrosion rates in operation with 30% by weight MEA. Table 2 shows the corrosion data and temperature at these positions at with a CO_2 concentration of 4% and 8% by volume in the flue gas, whilst Figure I shows the schematic of the positions (found in the Section *Appendix*).

Table 2: Corrosion rate and temperature on each position in pilot plant at 4% and 8% by volume CO_2 *(data retrieved from Kittel et al., 2009)*

	4% CO_2		8% CO_2	
	Corrosion Rate [μm.year^{-1}]	Temperature [°C]	Corrosion Rate [μm.year^{-1}]	Temperature [°C]
IP1: Stripper Overhead	535	103.91	538	100.44
IP2: Rich to Stripper	533	111.58	1075	110.99
IP3: Reflux and Vapour	83	26.23	233	28.42
IP4: CO_2 Product	22	26.32	23.6	35.54
IP5: Stripper Bottom	47	118.97	28	117.94
IP6: Absorber Bottom	4	49.16	5	46.08
IP7: Lean to Storage	2	54.18	2	54.95
IP8: Absorber Overhead	0	N/A	164	N/A
IP9: Absorber Offgas	49	50.16	698	53.44

This data indicates that the highest corrosivity is found at locations with the higher temperatures and fluid velocity, i.e., the inlet and outlet of the stripper. They also show that higher CO_2 in the flue gas causes higher corrosion rate.

The release of MEA and other amines is a significant concern in post-combustion capture due to amine loss in the absorber column. This concern is due to degradation reactions that generate toxic compounds like nitrosamines and nitramines (Luis, 2016; Poste, Grung and Wright, 2014; Baker and Lokhandwala, 2008). Nitrosamines have strong carcinogenicity and the foremost environmental concern regarding amine-based carbon capture is the risk of contamination by

nitrosamines (and nitramines but there is much less data pertaining to their toxicity) (Poste, Grung and Wright, 2014).

A study by Karl et al. (2011) indicates that toxicity to aquatic organisms is a "major concern" for the use of MEA as tolerable emissions were found to be only 12 tonnes/year. This coincides with results from a human and environmental impact assessment study which reported that scrubbing from carbon capture results in a "10-fold increase in toxic impacts on freshwaster" as opposed to a conventional power plant (Veltman, Singh and Hertwich, 2010).

Temperature and the concentration of SO_2 and O_2 have great effect on the degradation of MEA. This is shown by studies of kinetics conducted by Supap et al. (2009). The findings indicated that as the temperature and concentrations of MEA, O_2, and SO_2 rose, the rate of MEA degradation also increased. Conversely, an increase in CO_2 concentration had the opposite effect. A semi-empirical model of the degradation was developed to fit the experimental data shown in Equation 5 **(**Supap et al., 2009):

$$-r_{\mathrm{MEA}} = \{6.74 \times 10^9 \, e^{-(29,403/RT)} [\mathrm{MEA}]^{0.02}([\mathrm{O}]^{2.91} + [\mathrm{SO_2}]^{3.52})\}/\{1 + 1.18[\mathrm{CO_2}]^{0.18}\} \qquad (5)$$

As indicated by the higher order of reaction, SO_2 exhibits a greater tendency to induce degradation in MEA compared to O_2. As such, SO_2 must be separated before the flue gas enters the CO_2 stripper.

As the most used amine solvent for carbon capture, current research focuses on the minimisation of the high energy consumption during solvent regeneration of MEA. The energy requirement for an aqueous solution of 30% w/w has been reported to be 4215 kJ/kg-CO_2 (Bandyopadhyay, 2010) but can range between 3200 to 5500 kJ/kg-CO_2 (Luis, 2016), which are very high values.

Ammonia has been considered as an alternative due to its lower energy consumption for regeneration and lack of corrosion problems. The lower energy consumption (27% of that of MEA) calculation is shown in the following Table 3.

Table 3: Comparison of energy consumption of an MEA-based process and ammonia-based process *(using data from Bandyopadhyay, 2010)*

	MEA-based process [kJ/kg-CO_2]	Ammonia-based process [kJ/kg-CO_2]
Sensible heat	865	298
Reaction energy	1 920	644
Stripping stream	1 430	205
Total energy	4 215	1 147

Although the production of MEA per unit of mass is more energy intensive, the greater quantity of ammonia required results in a higher energy requirement for solvent production overall. Additionally, a large amount of CO_2 is released in the Haber-Bosch process, which results in a high amount of indirect emissions (Strube, Pellegrini and Manfrida, 2011). Thus, it is not a viable alternative to MEA.

An amine-based carbon capture also requires a great amount of water, in the cooling of the amines and to help compress the captured CO_2. Cooling the amines used for CO_2 absorption, which inherently generates heat, adds an extra burden on the cooling tower, resulting in increased water loss. Additionally, the compression of CO_2 to achieve the supercritical conditions necessary for storage also necessitates cooling. According to research conducted at NETL, which is operated by the U.S. Department of Energy, the incorporation of carbon-capture technology in the U.S. electricity sector by 2030 would result in an 80 percent increase in water consumption, equivalent to approximately 7500 megalitres per day. (Moore, 2010).

The energy required for regeneration and the water required for cooling counteract the benefits of capturing the carbon dioxide.

In conclusion, although it is currently the most employed and developed method of carbon capture, amine-based post-combustion carbon capture possesses problems including regeneration, degradation, and toxicity. However, amines are still widely used (particularly MEA) due to their selective and rapid reaction with CO_2.

3.1.2. Adsorption by Solid Sorbents

Research has been conducted on adsorption using solid solvents due to their potential to reduce the energy requirement of CO_2 capture as opposed to aqueous amine solvents, due to their low moisture content (Samanta et al. 2011; Dutcher, Fan and Russell, 2015). Additionally, adsorption generates no liquid waste (Boer, Langerak and Pescarmona, 2023). A variety of solid physisorbent materials can selectively adsorb CO_2 due to Van der Waals attraction between the adsorbent surface and the CO_2.

A good adsorbent ideally has the following (Boer, Langerak and Pescarmona, 2023):

- High adsorption capacity: high amount of CO_2 absorbed.
- High working capacity for CO_2: large difference between adsorption capacity at operating and regeneration pressure, ergo, lower amount of adsorbent required.
- Selective adsorption of CO_2: high selectivity towards CO_2 over other gases, leading to reduced capture costs.

At present, zeolites, and metal-organic frameworks (MOFs) are regarded as the most promising and extensively researched adsorbents.
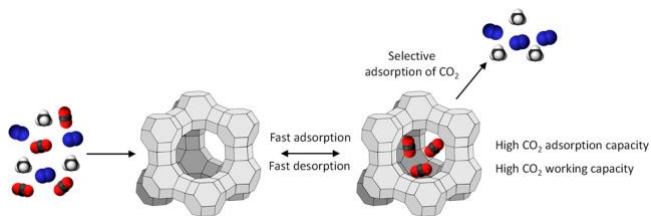
3.1.2.1. Zeolites

Figure 7: Schematic of CO_2 absorption by zeolites *(image retrieved from Boer, Langerak and Pescarmona, 2023)*

Zeolites are silicates made up of tetrahedral $[SiO_4]^{4-}$ interconnected by oxygen bridges. This arrangement creates three-dimensional crystalline structures with micropores, meaning the pores are less than two nanometres in diameter. Zeolites are often classified as aluminosilicates as a fraction of the silicon atoms can be substituted by other elements and aluminium is the most

common substituent (thus forming $[AlO_4]^{-5}$) (Boer, Langerak and Pescarmona, 2023; Garshasbi, Jahangiri and Anbia, 2017).

Zeolites vary in pore size, and for efficient CO_2 capture, the adsorbent should possess large volume and surface area of pores (0.5-0.7 nanometres) (Kumar, Srivastava and Koh, 2020).

A promising zeolite is Zeolite 13X, which is synthesised by hydrothermal treatment of natural clays. The natural clay used changes the suffix, for example 13X-B is prepared from bentonite, 13X-F is prepared from feldspar and 13X-K from kaolin. The Table 4 shows CO2 adsorption capacities for various Zeolite 13X formations.

Table 4: CO_2 adsorption capacities of different zeolite 13X formations at different temperatures (using data from Garshasbi, Jahangiri and Anbia, 2017)

Adsorbent	Temperature [K]	CO_2 adsorption capacity [mmol/g]
Zeolite 13X-B	298	4.9
Zeolite 13X-B	308	4.2
Zeolite 13X-K	298	6.9
Zeolite 13X-K	308	6.2
Zeolite 13X-F	298	3.9
Zeolite 13X-F	308	3.2

This study shows that Zeolite 13X-K is the most adsorptive formation and indicates that all Zeolite 13X formations possess lower CO_2 adsorption capacities at higher temperatures. Zeolite 13X can be formed from other clays and research continues investigating these zeolites.

3.1.2.2. Metal-Organic Frameworks

Metal-organic framework (MOF) materials are built up from metal ions and organic ligands, and their pore shape, size and chemical environment can be systematically designed. They have been reported to have very high gas uptake capacities. However, the majority of MOF materials are tested at the microgram scale and are yet to be tested at large scale (Babu et al., 2015; Qazvini, Babarao and Telfer, 2021).

3.1.2.3. Pressure Swing Adsorption and Temperature Swing Adsorption

CO_2 desorption using the adsorption method can be achieved by periodically adjusting temperature and pressure, as the amount of gas absorbed varies with alterations in these characteristics. As such, the adsorption separation method can be separated into pressure swing adsorption (PSA) and temperature swing adsorption (TSA) (Bi and Ju, 2022).

PSA occurs when the regeneration of the adsorbent is performed by reducing the total pressure the system, which results in the pressure ¨swinging¨ from high pressure in the feed to low pressure in regeneration. If the concentration of CO_2 is high, then PSA is preferred; if not, the process will take much longer, and TSA is preferred (Grande, 2012; Sifat and Haseli, 2019). In TSA, the adsorbents are heated to regenerate them (using steam, a hot inert gas or external heating). Due to this, TSA has high energy requirements and PSA is usually preferable (Meisen and Shuai, 1997). The two methods are shown in Figure 8, showing the flue gas entering beds of adsorbents and the CO_2 released to the condensate for TSA or to the adsorbed gas in PSA.
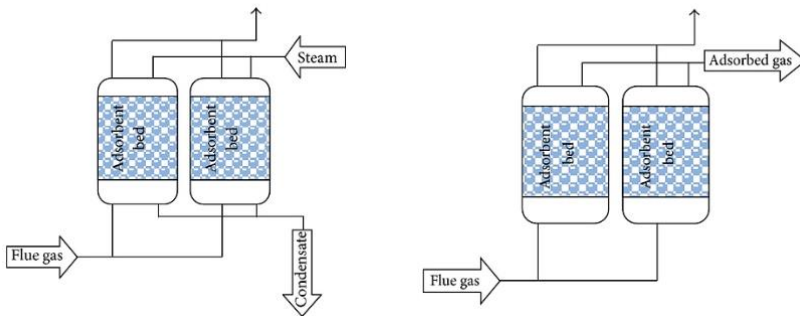

Figure 8: Schematic diagram of TSA using heating by steam (left) and PSA (right) adsorption regeneration cycles (Songolzadeh et al., 2014)

3.1.3. Membrane Separation

Membrane technology is a physical separation process that involves the use of a semipermeable barrier to separate gas mixtures containing two or more components. This barrier allows some components to pass through, creating a permeate stream, while retaining the remaining components in a separate stream called the retentate (Khalilpour et al., 2015).

The major advantages of membrane systems over absorption are reduced equipment sizes, lower energy requirements and the absence of waste streams. (Boer, Langerak and Pescarmona, 2023)

Additionally, membrane systems are particularly advantageous for high concentration of CO_2 with low flow rates. (Baker and Lokhandwala, 2008). This charecteristic is shown in Figure 9, where the schematic indicates the region of membrane preference located in the bottom right quadrant. This region corresponds to conditions of elevated CO_2 concentration and reduced gas flow rate. However, the specific diagram will vary based on the site and must be customised for each case.
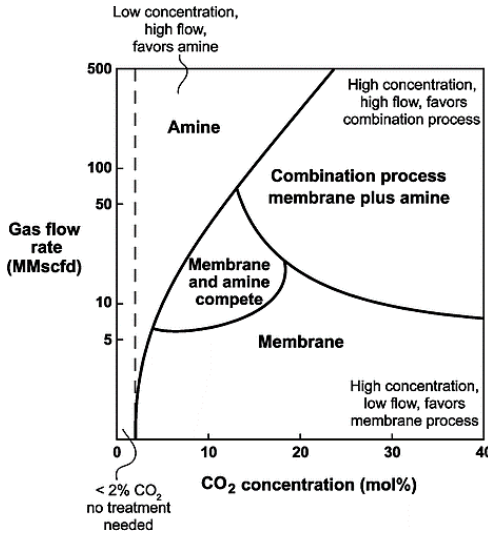

Figure 9: Schematic plot illustrating the effect of gas flow rate and carbon dioxide concentration in the gas on the choice of carbon dioxide removal technology (Baker and Lokhandwala, 2008)

Gas separation is characterised by the selectivity (α) and the gas permeance (P) of the individual components. The permeance of component i (P_i) is calculated as follows:

$$P_i = \frac{N_i}{\Delta p_i \cdot A} = \frac{F_i}{\Delta p_i} \tag{6}$$

In Equation 6, the molar flux of compound i (F_i) is represented by the permeate rate of component i (N_i) in terms of moles per second (mol s^{-1}), with A denoting the membrane area and Δp_i indicating the partial pressure difference of component i across the membrane. The SI

unit for permeance is mol s^{-1} Pa^{-1}, but is represented commonly in GPUs, where GPU = 3.3928 x 10^{-10} mol s^{-1} Pa^{-1} (Senior, Morris, and Lewandowski, 2013).

The pure gas selectivity is calculated as the ratio of the permeance of the faster permeating component (H$_2$) to the permeance of the less permeating component (CO$_2$), as shown in Equation 7:

$$\alpha = \frac{P_{H_2}}{P_{co_2}} \qquad (7)$$

In membrane technology, achieving high permeability is crucial to reduce both the membrane surface area and the separation time. On the other hand, high selectivity is necessary to obtain optimal purity**.** However, one of the major hurdles in membrane technology is finding the right balance between permeability and selectivity, as they often present a trade-off.

An analysis conducted by Zhai and Rubin (2013) showed that a higher permeance leads to a lower CO$_2$ avoided cost, resulting in overall lower costs. Avoided CO2 cost is covered in Section 5.2.2. A two-stage membrane for 90% carbon capture was tested at 0.2 bar. Figure 10 below shows that a higher permeance results in less costs and the difference is more pronounced with higher membrane module prices.
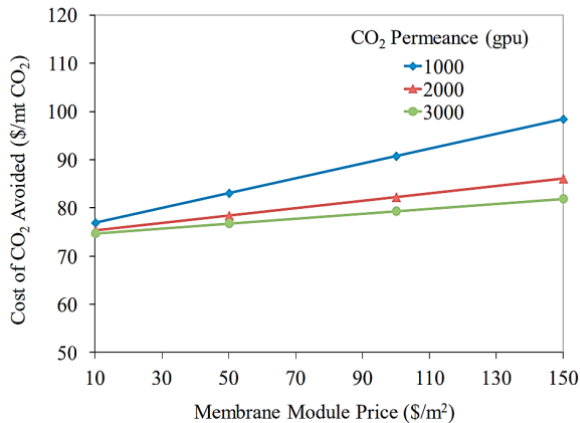

Figure 10: Effects of CO$_2$ permeance and membrane module price on cost of CO$_2$ avoided
(image retrieved from Zhai and Rubin, 2013)

Conversely, membranes suffer many issues including low selectivity at high-pressure operation and low high-temperature stability (the latter can lead to inadequate performance) (Basile et al., 2011; Lei et al., 2020).

3.2. PRE-COMBUSTION CARBON CAPTURE

Pre-combustion carbon capture is the removal of carbon dioxide before combustion and is commonly used in integrated gasification combined cycle power plants, which rely on gasification for power generation.

Pre-combustion power plants separate synthetic gas (known as syngas), which is a mixture of predominantly H_2 and CO. This can be produced by steam reforming or gasification. Steam reforming is undergone in natural gas power plants, typically with methane, and has the following reaction (Sifat and Haseli, 2019):

$$C_xH_y + xH_2O \rightarrow xCO + \left(x + \frac{y}{2}\right)H_2 \quad (8)$$
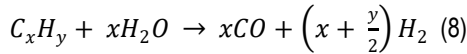
Gasification is another method to obtain syngas and is the process of converting carbonaceous raw material such as coal into syngas. This takes place in a gasifier, which is typically a high-pressure and high-temperature vessel. In this process, a hydrocarbon feed material is fed with pure oxygen, resulting in the partial oxidation of the hydrocarbon (Theo et al., 2016). Typically, the oxygen is supplied by an air seapration unit. The reaction is shown in Equation 9, as presented by Sifat and Haseli (2019).

$$C_xH_y + \frac{x}{2}O_2 \rightarrow xCO + \left(\frac{y}{2}\right)H_2 \quad (9)$$
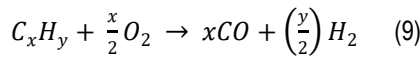
The syngas is then cooled and sent to a particulate removal unit, typically by cyclone separation. The gas is then shifted by adding steam and reacting over a catalyst in a water gas shift reactor (Theo et al., 2016). As demonstrated by Sifat and Haseli (2019), this reaction yields hydrogen gas and carbon dioxide, as indicated in Equation 10.

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad (10)$$
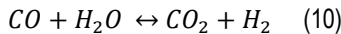
After leaving the water shift gas reactor, the CO_2 concentration is in the rage of 15-60% (dry basis) and a total pressure of 2-7 MPa, both of which are much higher than in post-combustion carbon capture. As such, there is an increased driving force for separation as the CO_2 has a higher partial pressure, and the carbon dioxide can be efficiently captured from syngas, which can then be transported to be utilised or safely stored (Figueroa et al., 2008; Wang & Song, 2020). The Figure 11 below shows a simplified schematic of a pre-combustion carbon capture system.
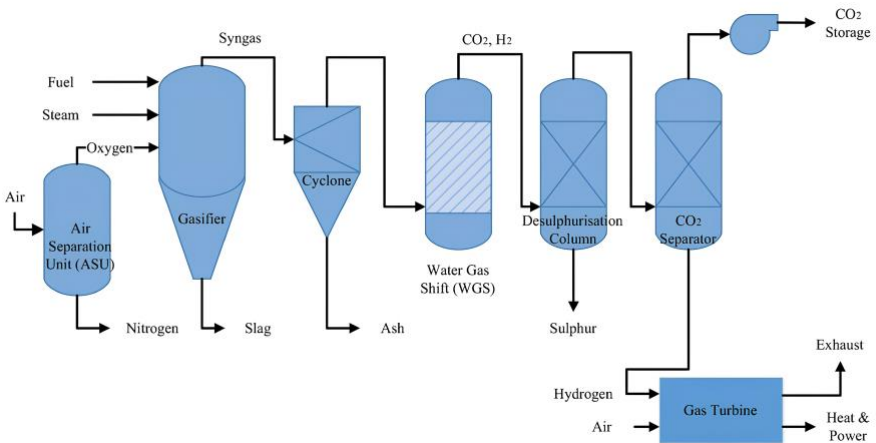

Figure 11: Process flow diagram of pre-combustion carbon capture system with gasification
(image retrieved from Theo et al., 2016)

Due to the increased driving force, a much lower volume of gas needs to be treated. Moreover, theoretically, the greater efficiency results in a lower energy demand for carbon capture and compression operation as opposed to post-combustion capture (Theo et al., 2016).

Despite this, pre-combustion carbon capture is more complex than post-combustion carbon capture, leading to a higher investment cost. Particularly gasifiers have low availability and poor operational availability, (Mondal, Balsora and Varshney, 2012).

3.2.1. Absorption with Chemical Solvents

As with post-combustion, aqueous amine solutions are used as solvents and they possess similar negatives and benefits. For pre-combustion carbon capture, MDEA is a widely used

solvent due to the high pressure, high CO_2 concentration gaseous stream of the method (Antonini et al., 2021).

3.2.2. Absorption with Physical Solvents

Unlike post-combustion carbon capture, most commercially developed technologies of pre-combustion carbon capture use physical solvents. If carbon dioxide is under great pressure (> 10 bar) as in pre-combustion, physical solvents have a larger absorption limit and are used to capture CO_2. They are very dependent on temperature and pressure, and the flue gas from post-combustion is not in a suitable range. (Chen, 2022; Songolzadeh et al., 2014).

The physical absorption method functions by altering the operating pressure and temperature between CO_2 and the absorbent to facilitate the absorption and desorption of CO_2. For regeneration (desorption), the solvent is flashed to atmospheric pressure or vacuum. The absorbent does not react with CO_2. (Bi and Ju, 2022; Burr and Lyddon, 2008).

Existing physical solvents processes include Selexol, Rectisol and Purisol. Selexol uses a mixture of dimethyl ether and propylene glycol as the solvent, Rectisol uses chilled methanol (at approximately - 35°C to − 75°C) and Purisol uses n-methyl-2-pyrrolidone (at ambient temperature or up to -15°C) (Chen, 2022). The advantages and disadvantages of each method (in the context of carbon capture) obtained from a literature review are illustrated in Table 5.

Table 5: Summary of the advantages and disadvantages of the principal physical absorption processes

Method	Advantage	Disadvantage	Reference
Selexol process	■ Very little to no energy required for regeneration. ■ Very low vapour pressure (0.00073 mmHg at 25 °C)[a]. ■ Degradation does not occur.	■ High solvent costs. ■ High CO_2 partial pressure required. ■ High viscosity (5.8 cP at 25 °C)[a].	(Burr and Lyddon, 2008), (Ghasem, 2020), (Sifat and Haseli, 2019)
Rectisol process	■ High thermal and chemical stability.	■ High vapour pressure (125 mmHg at 25 °C)[a].	(Ghasem, 2020), (Sifat and Haseli, 2019)

	■ No reboiler required as there is no degradation, and no regeneration heat is required. ■ Low solvent cost. ■ Low solvent flow rate for CO_2 removal.	■ High capital and operating cost due to refrigeration requirement of solvent.	
Purisol process	■ Very effective for high-pressure, highly CO_2 concentrated gas. ■ High selectivity for H_2S.	■ Relatively high vapour pressure (0.4 mmHg at 25 °C)[a]. ■ Requirement of water scrubbing to avoid excessive solvent loss.	(Burr and Lyddon, 2008)

a — Data retrieved from Burr and Lyddon, 2018

A study by Park et al. (2015) compared these three physical solvents for a two-stage pre-combustion CO_2 capture process in ASPEN Plus. Critically, the energy consumption at different CO_2 capture percentages was simulated for each solvent. The results are shown in Figure 12, indicating that higher CO_2 necessitates higher energy consumption and that Selexol is the most efficient followed by Purisol and Rectisol.
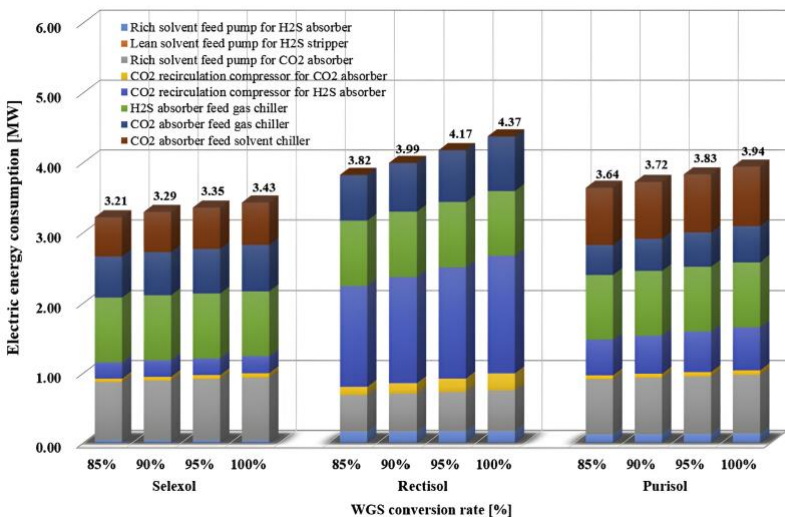
Figure 12: Calculation result of electric energy consumption depending on CO_2 capture percentages for each solvent. The legend shows the proportions of electrical consumption for each process. *(image retrieved from Park et al., 2015)*

The disadvantages of physical solvents are their low adsorption capacity and selectivity for CO_2 separation, unlike amines (chemical absorbents), which react rapidly, selectively, and reversibly with CO_2 (Songolzadeh et al., 2014).

3.2.3. Cryogenic Separation

Cryogenic CO_2 separation is a physical process in pilot development where CO_2 is subjected to extremely low temperatures, either causing it to condense or sublimate. By subjecting the mixed gas to multiple rounds of compression and cooling, the phase of CO_2 is altered, causing the separation of CO_2 from the rest of the gas mixture (Bi and Ju, 2022; Songolzadeh et al., 2014).

Cryogenic separation has many advantages, including a direct production of liquid CO_2 (thus avoiding compression) and an ability to easily scale-up the process to industrial scale. Moreover, the technology lacks solvents and avoids the disadvantages including corrosion and water consumption (Songolzadeh et al., 2014).

It is used for streams with high concentrations of CO_2, as dilute streams require a large amount of energy, which renders the process uneconomical (Mondal, Balsora and Varshney, 2012). For this reason, the process is not generally used for post-combustion carbon capture.

An accurate thermodynamic phase study of the gaseous mixtures is imperative for designing an efficient and economical cryogenic CO_2 capture process, as this helps generate the pressure and temperature conditions for the bubble and dew point curves, 3–phase locus, and CO_2 freeze–out line for the CO_2 gaseous mixtures. The increasing demand for cryogenic separation has pushed researchers to obtain more precise thermodynamic data. (Babar et al., 2019). The following Figure 13 shows the effect of CO_2 concentration on the pressure and temperature requirements to condense or sublimate CO_2.
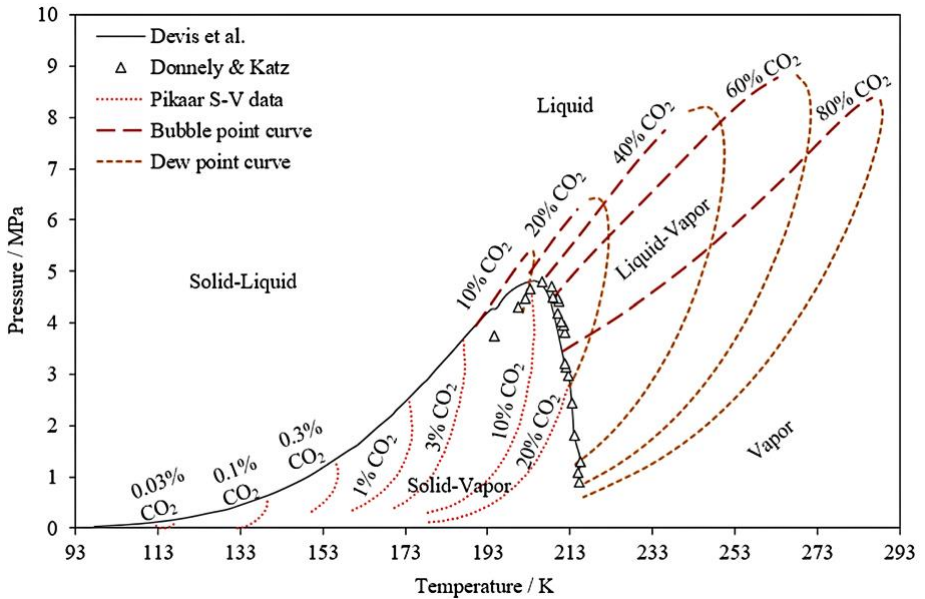
Figure 13: Effect of CO_2 concentration on P-T phase envelope. Devis et al., Donney & Katz, Pikaar S-V data refers to data from other authors, which contributed to the above image
(image retrieved from Babar et al., 2019)

A major consideration for cryogenic separation is the presence of water vapor in the CO_2 feed mixture, which can lead to the formation of solid CO_2 clathrates and ice causing major plugging problems. Consequently, a costly process is required to remove all water traces in the CO_2 feed (Mondal, Balsora and Varshney, 2012).

Additionally, cryogenic separation has a huge energy demand to reach the low temperatures required. Clodic et al. (2005) stated the total energy varied between 647.7 and 1 248.6 kJ/kg CO2 depending on the cooling system's efficiency. In the majority of research studies, the main objective for optimization centers around the reduction of energy consumption. (Bi and Ju, 2022).

Cryogenic separation is in a pilot stage of development and is a promising separation method for pre-combustion (and oxy-fuel combustion) carbon capture.

3.2.4. Additional Studies

As a novel separation technique, there have been many studies investigating pre-combustion carbon capture. Table 6 below shows additional important investigations into pre-combustion carbon capture technology.

Table 6: Literature review of recent studies pertaining to pre-combustion carbon capture

Year	Method of Separation	Conclusions	Reference
2010	Physical-chemical absorption (using MDEA)	■ CO_2 capture using MDEA was tested and compared to reference plant. ■ Net plant efficiency (with CO_2 capture) was 50.65% (8% less than reference combustion cycle without capture), CO_2 capture removal was 91.6%. ■ Huge loss of efficiency (17%) can be predicted if all detrimental effects of CO_2 are considered.	(Romano, Chiesa and Lozza, 2010)
2011	Adsorption (using activated carbon)[a]	■ Study of CO_2 equilibrium adsorption capacity and breakthrough time using activated carbon. ■ Partial pressure of CO_2 was found most significant as carbon capture capacity and breakthrough time were directly proportional to it and inversely proportional to temperature.	(García et al., 2011)
2013	Absorption (using aMDEA)[b]	■ Pilot plant of 14 MWth, captured 1000 tonnes of CO_2 and produced 6 tonnes of H_2 in 550 hours of operation. ■ Technical assessment shows improvement possible in efficiency in operation and integration with IGCC process.	(Burr and Lyddon, 2008)
2014	Absorption (using MDEA)	■ Assessment of IGCC power plants with air-blown gasification (as opposed to typical oxygen-blown gasification). ■ Results showed competitive efficiency penalty and carbon to typical oxygen-blown gasification.	(Moioli et al., 2014)

| 2018 | Adsorption (using LDOs)[c] | ■ Pilot-scale test system using LDOs shown to be successful with 1089 h of accumulated operation and 75 h of continuous operation, sustaining a CO_2 removal ratio higher than 91.7%. | (Zhu et al., 2018) |
| | | ■ It was a 4-column elevated temperature (250-400°C) PSA system with processing capacity of 4.0-6.6 Nm^3 h^{-1}. | |

a — Chosen activated carbon was Norit R2030CO₂ (García et al., 2011).

b — aMDEA refers to activated MDEA using piperazine, which improves reactivity of MDEA thus reducing heat of reaction and circulation rate requirements for CO_2 removal (lowering reboiler energy requirements as a result) (Burr and Lyddon, 2008).

c — LDOs are potassium-promoted Mg–Al layered double oxides (Zhu et al., 2018).

This literature review reveals that many new separation methods are being investigated for pre-combustion carbon capture. A very common separation method is absorption with MDEA and its variants. Generally, the studies show a positive outlook for pre-combustion capture, however, like post-combustion carbon capture, there are great efficiency losses, which are likely greater than previously expected.

3.3. OXY-FUEL COMBUSTION CARBON CAPTURE

Oxy-fuel combustion is a novel but promising technology that can be used to capture and sequester carbon dioxide from existing plants. It is a type of post-combustion carbon technology, as the CO_2 is captured after combustion. The idea for oxy-fuel combustion was originally proposed by Abraham in 1982 and pertained to providing a CO_2-rich flue gas for enhanced oil recovery, conceived as a retrofit option for coal-fired plants (Senior, Morris and Lewandowski, 2013).

While conventional combustion employs air as the oxidant, oxy-fuel combustion involves the use of either pure oxygen or a mixture of pure oxygen and carbon dioxide in the combustion reaction. As there is little to no nitrogen gas, the flue gas consists primarily of carbon dioxide and water, which means that no solvent is required. Some fraction of flue gas is recycled (which is referred to as recycled flue gas) to lower the flame temperature**.** A condensation process removes the water, so the almost pure CO_2 can be captured. Whereas in conventional air, condensation

starts at 80 °C, in oxy-fuel it takes place at 150 °C. (Yadav and Mondal, 2022; Stanger et al., 2015).

Oxy-fuel combustion is generally composed of four units: air separation unit (ASU) to produce oxygen, a boiler for the combustion of fuel, a flue gas processing unit for flue gas cleaning and finally, a CO_2 processing unit for final purification of the carbon dioxide to transport and store it (Stanger et al., 2015). Figure 14 shows a schematic of the process. The figure also illustrates the potential utilization of steam from the boiler in a steam turbine, however this is outside the scope of this project.
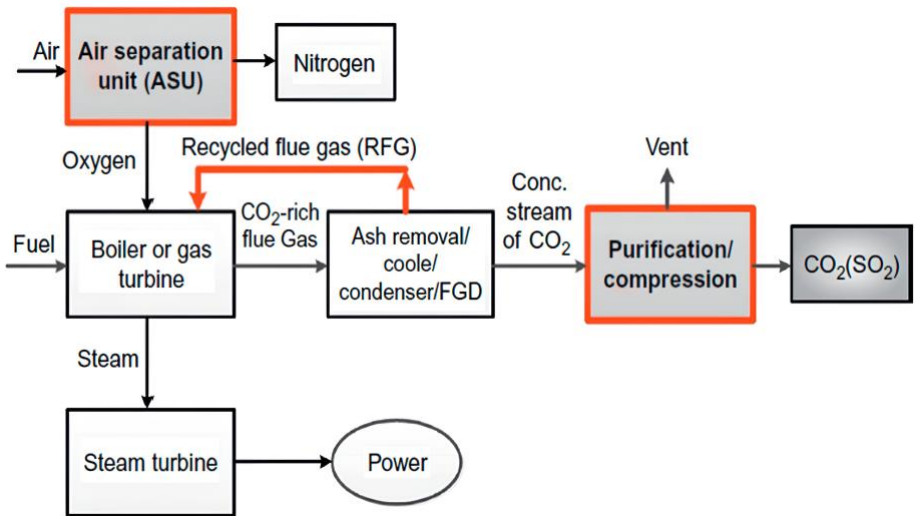
Figure 14: Schematic of an oxy-fuel combustion system *(image retrieved from Sifat and Haseli, 2019)*

An air separation unit (ASU) is required to provide purified oxygen to the steam generator island. According to Thimsen, Wheeldon and Dillon (2011), cryogenic distillation of oxygen from air is the only technology "sufficiently mature" for a commercially viable oxy-fuel power plant. A cryogenic ASU includes raw air filtration, air compressors, water vapor and trace CO_2 removal, pre-refrigeration and a cryogenic cold box which includes heat exchanges, an oxygen-nitrogen distillation column and an expansion turbine (Thimsen, Wheeldon and Dillon, 2011).

The ASUs for oxy-fuel combustion differ from those producing industrial oxygen (at 99.5% purity) as summarised in the Table 7 below:

Table 7: Summary of differences between ASU unit requirements for typical industrial oxygen and for oxy-fuel combustion

Topic	Industrial Oxygen Production	Oxygen for Oxy-fuel Combustion	Reference
Oxygen production requirements	■ Typical ASU's have much lower oxygen production. ■ Largest ASU has total production capacity of 5 000 tonnes/day (an Air Liquide facility located in South Africa)	■ Multiple ASU units are necessary. ■ Oxygen requirement of 20 000 tonnes/day for 800 MW plant and 10 000 tonnes/day for 500 MW plant.	(Allam et al., 2003), (Air Liquide, 2021), (Thimsen, Wheeldon and Dillon, 2011)
Pressure	■ Industrial oxygen is delivered at high pressure.	■ Oxygen to an oxy-fuel combustor is fed at low pressure (~ 2 bar).	(Thimsen, Wheeldon and Dillon, 2011)
Purity	■ Industrial oxygen and medical oxygen are commonly produced at 99.5% purity.	■ 95-97% purity is acceptable for oxy-fuel; achievable without costly oxygen-argon separation[a], thus reducing energy consumption by 10%.	(Šulc & Ditl, 2021), (Thimsen, Wheeldon and Dillon, 2011)

a —— Separating oxygen and argon is more complex than separating oxygen and nitrogen as there is an unfavourable vapour liquid equilibrium due to the close boiling temperatures (Šulc & Ditl, 2021).

The oxygen requirements for oxy-fuel combustion means conventional ASUs can't be used and multiple ASU units are necessary.

In a conventional power-plant, nitrogen in the air acts as a temperature moderator. As such, in oxy-fuel combustion, the absence of nitrogen in the combustor leads to excessively high flame temperatures. A solution to control and maintain the temperature within an acceptable range is to introduce recycled CO_2 into the combustor along with pure oxygen. Alternatively, injecting steam into the combustion chamber is another approach (Sifat and Haseli, 2019).

As shown in Figure 14, the flue gas is to be treated in a flue gas desulphurisation unit (FGD) where all sulphur products are removed.

Unlike in post-combustion and pre-combustion, the CO_2 is captured by condensing steam, as the combustion products are mainly CO_2 and water. This means that there is no need for the

separation techniques presented in the previous sections (absorption, adsorption etc.), and the process is, in theory, the most economically viable (the ASU is the biggest challenge for this technology). However, cryogenic separation can also be used in oxy-fuel combustion method due to the purity of the flue gas (Songolzadeh et al., 2014).

As in the other carbon capture methods, the CO_2 must be compressed to be transported and stored.

3.3.1. Additional Studies

Like pre-combustion carbon capture technology, oxy-fuel carbon capture is a novel process. There are many studies being conducted on oxy-fuel carbon capture.

Table 8: Literature review of recent studies pertaining to oxy-fuel combustion carbon capture

Year	Remarks	Reference
2011	■ Study of Total 30MWth plant. Flames from burners were visualized by endoscope technique. ■ Showed that a lower flue gas recirculation resulted in longer flames. ■ Showed inverse relationship between flue gas recirculation and NO_x emissions. Alternatively, a linear relationship existed between excess oxygen and NO_x emissions.[a]	(Marcano et al., 2011)
2011	■ Study of effect of sulfur in oxy-fuel reactor. ■ Was found that oxy-fuel with recycle stream lead to lower volume of flue gas to treat but also significantly higher concentrations of impurities (particularly SO_x) in furnace.	(Stanger and Wall, 2011)
2012	■ Study of ETR using blends of coal under various oxy-fuel atmospheres. ■ Showed that replacing N_2 with O_2 caused increase of ignition temperature. ■ Found that emissions of NO during oxy-fuel combustion were lower than under air-firing but remained similar as O_2 increased in concentration (for oxy-fuel).	(Riaza et al., 2012)

a – These relationships are shown in Figures II and III in the Section *Appendix*.

4. TRANSPORT AND STORAGE

4.1. TRANSPORT

Presently, most projects opt for a combined approach of developing transport and storage systems. CO_2 transport infrastructure moves the CO_2 from point of capture to point of storage (or utilisation). Pipelines and ships offer the highest scalability and the lowest cost per tonne of CO_2 for its transport (International Energy Agency, 2022a). Whereas ship transport is preferable for long distances (>1500 km), transportation by pipelines will occur in most cases (Bui et al., 2018).

In pipelines, the volume of CO_2 is reduced by transporting at a high pressure of around 80 MPa (Doctor and Palmer, 2005) but pressures of 110 MPa have been found in literature. As aforementioned, this requires more energy for post-combustion capture processes as the resulting CO_2 needs to be pressurised from atmospheric pressure. The expenses associated with transporting CO_2 through pipelines can vary significantly and rely on factors such as the length of the pipeline, its diameter, the terrain it crosses, and the chosen route.

According to Göttlicher and Pruschek (1997), the energy requirement for liquefaction by intercooled 5-stage compression from 1 bar (the pressure of CO_2 captured from post-combustion capture) to a pipeline pressure of 110 bar would require approximately 0.12 MWh/tonne-CO_2.

A techno-economic study conducted by Wang et al. (2010) demonstrated that an oxy-fuel plant with a capture efficiency of 90%, capturing 6057 tonnes of CO_2 per day would require 111 kWh per tonne of CO_2 for compression.

An analysis done by Petrescu and Calin-Cristian Cormos (2017) showed of an IGCC plant using pre-combustion carbon capture analysed the electricity cost and CO_2 losses to transport CO_2. This was done through simulation using the program ChemCAD process simulator. The pipeline distance was 800 km with 8 compressors each with 100 km between them, resulting in a pressure drop of 0.06 bar/km. The CO_2 is transported at 120 bar and 35 °C, in the supercritical region.

Table 9: Summary of electricity requirements and CO_2 losses for a simulated pre-combustion plant *(data retrieved from Petrescu and Calin-Cristian Cormos, 2017)*

Attribute	Value	Units
Electricity for compressors	0.1111	[MWh/tonne-CO_2]
CO_2 losses pipeline	0.407	[kg CO_2/MWh]
CO_2 losses compressors	0.305	[kg CO_2/MWh]

The simulated results indicate that for pre-combustion, the electricity to compress CO_2 to 120 bar (0.1111 MWh/tonne-CO_2) is less than the electricity required to compress from 1 bar to 110 bar in post-combustion. However, a direct comparison with the same pipeline pressure is required to accurately compare them. The results also show that the CO_2 losses are similar for the pipeline as for the compressors.

In general, pre-combustion capture has the lowest energy requirement for compression. This is further shown in Section 5.2.2., in an analysis conducted by the Massachusetts Institute of Technology (MIT) (2007).

4.2. STORAGE

The most significant carbon capture storage option are aquifers which are geological formations containing brine in porous rock.

Carbon dioxide can be pumped to depth of over one kilometre where the CO_2 is pressurised to a density of 200-800kg/m³. When CO_2 is injected into an aquifer, it displaces brine and creates a plume that typically ascends towards the top of the aquifer. At the interface between CO_2 and brine, a certain amount of CO_2 will dissolve in the brine, with a solubility of approximately 1-2% and some water will dissolve within the CO_2 plume. Over tens of thousands to millions of years, the CO_2 can mineralise into rock (United Nations Economic Commission for Europe, 2021). The Figure 15 below shows a simplified schematic of the process, showing the injection of CO_2 under the caprock. It also shows that the is CO_2 is most concentrated at the top region of the plume.
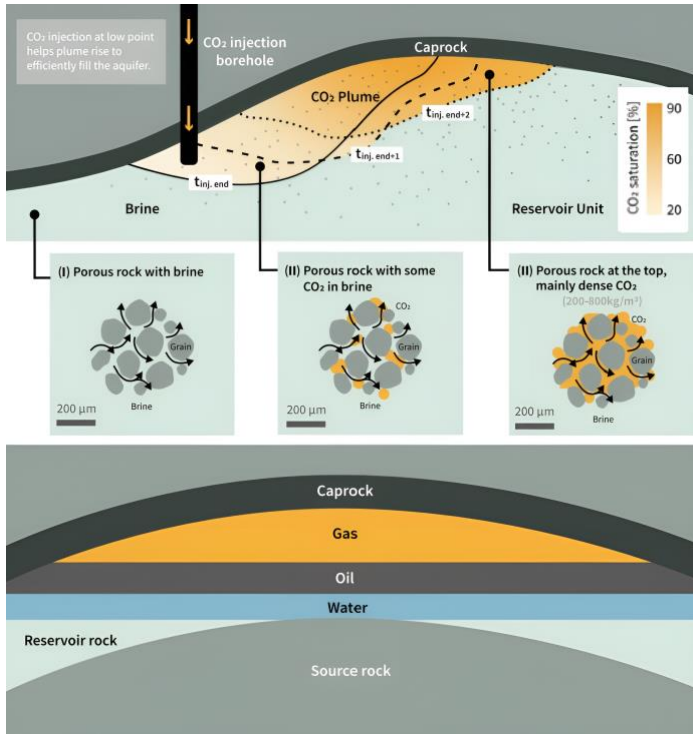
Figure 15: Simplified view of aquifer with a plume of CO₂ injected below a caprock *(image retrieved from United Nations Economic Commission for Europe, 2021)*

However, this process increases the acidity affecting the biome in the aquifer. Additionally, leakages of CO_2 or brine into sources of drinking water or soil can lead to detrimental consequences by lowering their pH. Furthermore, continuous leakages of CO_2 counteract the benefits of carbon capture for climate change mitigation. Leak-proof storage of CO_2 is impossible to guarantee (Robertson and Mousavian, 2022) and according to a sensitivity analysis done by Viebahn et al. (2007), the smallest thinkable leakage rate (0.0001%/annum) would cause a 31% increase of CO_2 emissions and a rate of 0.1%/annum would result in all CO_2 stored underground to be released within the next 6000 years. The following Figure 16 demonstrates this in a sensitivity analysis showing the effects of different storage leakage rates in the middle-term (between 0 to 10 000 years) and the long term (> 10 000 years).
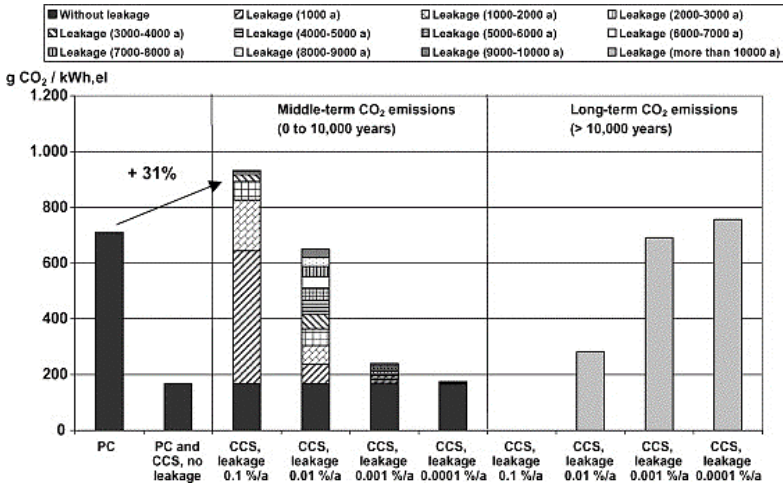
Figure 16: Comparison of middle-term and long-term release of carbon dioxide from capture from pulverised coal power plant. The y axis shows the grams of CO2 emitted per kWh of electricity produced. The x axis shows the leakage rates for each simulation. As shown in the legend, the different patterns represent different passages of time. 'a' refers to annum and 'el' refers to electricity production *(image retrieved from Viebahn et al., 2007)*

An analysis by Zahasky and Krevor (2020) indicated that 2700 gigatonnes of storage is required to meet the most ambitious climate change mitigation targets. Another analysis by Bui et al. (2018) showed that capping atmospheric CO_2 levels to ~450ppm would require the storage of 120-160 gigatonnes of CO_2 at a rate of about 10 gigatonnes per annum, achieved by 2050. Furthermore, the analysis posited that the storage requirement for 2100 could be as high as 1200-3300 gigatonnes of CO_2. However, an analysis by Zhang, Jackson and Krevor (2022) estimated that only 197 megatonnes of CO_2 was geologically sequestered over the period of 1996-2020, which falls far short of the requirements for climate change mitigation goals.

Ultimately, the high storage requirement and possibility of leakage are significant drawbacks to carbon capture's ability to abate climate change.

4.3. ENHANCED OIL RECOVERY

In addition to being stored, captured CO_2 can be utilised by pumping it into depleted oil and gas fields. This process is called Enhanced Oil Recovery (EOR), and although it can be done with

other gases and long-chain polymers, CO_2 is commonly used for its low cost and wide availability (Cuéllar-Franca and Azapagic, 2015; Integrated Flow Solutions, 2019)**.**

The aim is to recover additional amounts of oil from reservoirs that has experienced a decline in oil production below critical levels. The carbon dioxide is normally injected at a supercritical state, where the temperature-pressure conditions in the reservoir are above minimum miscibility pressure so that the injected CO_2 is fully miscible with the oil phase in the reservoir. This reduces the viscosity of the oil which is then displaced from the rock pores (Bui et al., 2018). Figure 17 shows a schematic of the process where the CO_2 is injected under the caprock, causing the oil and CO_2 to rise to the surface to be transported away.
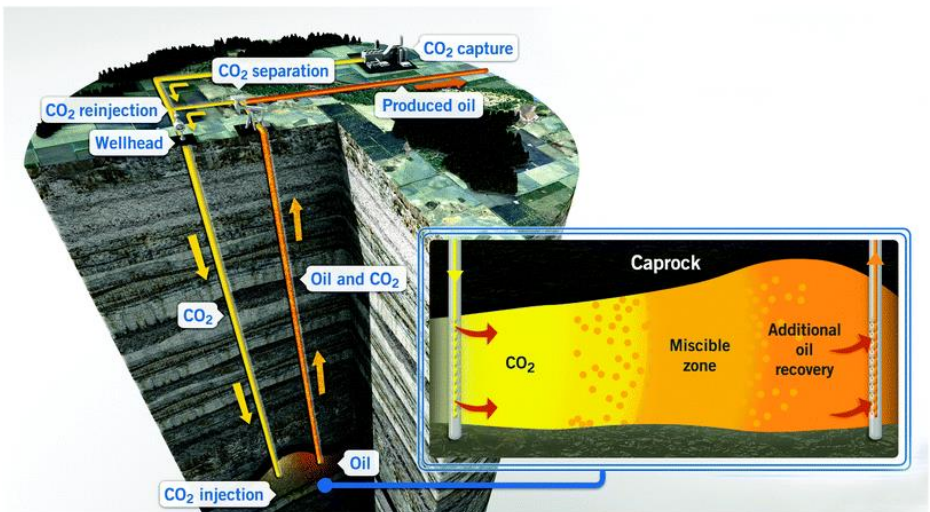
Figure 17: Schematic diagram of the CO_2-EOR process (image retrieved from Bui et al., 2018)

Additionally, the CO_2 can be injected permanently in the reservoir rather than pumping it back to the surface. In this case, the process is carbon capture and storage enhanced oil recovery (CCS-EOR) (Bui et al., 2018).

Although newer developments prioritise storage of carbon storage, currently enhanced oil recovery is the primary destination for captured CO_2 (International Energy Agency, 2022a).

In 2022, enhanced oil recovery comprises 73% of the total capacity of the current capture capacity with geological storage comprising of 27% (Robertson and Mousavian, 2022). This is shown in Table 10. The Global CCS Institute states out of the 30 operational CCS facilities, 9 are used for dedicated geological storage and 20 are used for enhanced oil recovery. The remaining facility is the Boundary Dam Carbon Capture and Storage Facility, which is used for both purposes (Steyn et al., 2022).

Table 10: Comparison of Enhanced Oil Recovery and Dedicated Geological Storage in terms of accumulated CO_2 capture and share of 2022 CO_2 capture amount (data retrieved from Robertson and Mousavian, 2022)

Carbon Capture Type	Accumulated Capture of CO_2 [Million Tonnes]	Share of 2022 39 MTPA[a] Capture [%]
Enhanced Oil Recovery	>240	~73%
Dedicated Geological Storage	<60	~27%
Total	~300	100%

a – MTPA refers to Million Tonnes per Annum

Enhanced oil recovery projects offset the benefits of capturing CO_2 as it facilitates the use of more fossil fuels. The net emissions for enhanced oil recovery depends on how much CO_2 is stored. However, an analysis by the International Energy Agency (2015) reveals that the current EOR approach with a representative net utilisation of 0.3 tCO_2/bbl (tonne per barrel) would result in net emissions of 0.8 tCO_2-eq per tonne of CO_2 delivered if the recovered oil is combusted. The only way for there to be negative net emissions is if the utilisation is greater than ~0.6 tCO_2/bbl. Nevertheless, the use of captured CO_2 in enhanced oil recovery will always offset the emissions mitigated and currently will lead to positive net emissions when the oil is combusted.

If enhanced oil recovery is an option for storage, the following operation costs for each process are estimated. CO_2 separation from exhausting gases is between 24 to 52 €/tonne-CO_2, transportation to storage location: 1 to 6 €/tonne CO_2 per 100 km, and storage: −28 to 42 €/tonne CO_2. As aforementioned, the CO_2 capture is the most costly process. The negative value for storage is a result of economic benefit with EOR (Songolzadeh et al., 2014).

In summary, although Enhanced Oil Recovery (EOR) holds economic appeal, it undermines the advantages of carbon capture in mitigating climate change.

5. COMPARISON OF CARBON CAPTURE TECHNOLOGIES

5.1. COMPARISON OF METHODS OF SEPARATION

Whereas chemical absorption methods (aqueous amine solutions) and physical absorption methods are mature technology with wide applications, adsorption, membrane separation and cryogenic separation are all new technologies with limited applications (Bi and Ju, 2022). The following Table 11 summarises the main advantages and disadvantages of these separation methods, obtained by bibliographic review.

Table 11: Summary of the advantages and disadvantages of separation methods for post-combustion carbon capture.

Method	Advantage	Disadvantage	Scale	Reference
Chemical Absorption (MEA, DEA and other amine solutions)	■ Mature and developed technology. ■ Reacts rapidly. ■ Reversible process. ■ Inexpensive solvent.	■ Produces corrosive and toxic degradation products. ■ High regeneration energy requirement.	Industrial	(Bi and Ju, 2022), (Songolzadeh et al., 2014)
Physical Absorption (Selexol, Rectisol, Purisol)	■ Mature and developed technology. ■ Low solvent regeneration requirement. ■ Low corrosivity.	■ Dependent on temperature and pressure (unsuitable for post-combustion). ■ Low absorption capacity and selectivity for CO2 separation.	Industrial	(Burr and Lyddon, 2008), (Songolzadeh et al., 2014)
Adsorption (Solid Solvents)	■ Lower regeneration energy. ■ No liquid waste.	■ Low selectivity at high-pressure. ■ Low high-temperature stability.	Pilot	(Basile et al., 2011), (Boer, Langerak and Pescarmona, 2023), (Lei et al., 2020),

		Low adsorption capacity.		(Songolzadeh et al., 2014)
Membrane separation	Effective for high CO_2, low flow. Reduced equipment size.	High energy cost.	Experimental	(Baker and Lokhandwala, 2008), (Boer, Langerak and Pescarmona, 2023)
Cryogenic separation	No chemical absorbent required. Good scalability. Liquid CO_2 achieved directly.	High energy cost. Costly water removal process required. Unsuitable for post-combustion.	Pilot	(Babar et al., 2019), (Mondal, Balsora and Varshney, 2012)

Despite the advantages, the separation techniques in experimental and pilot development stages require further research and testing to show their viability, particularly at industrial scale.

5.2. COMPARISON OF THE THREE TYPES OF CARBON CAPTURE

Table 12 shows a summarised comparison of the three types of carbon capture. The limitation of the following comparison is that the true efficiencies of the processes can only truly be investigated at industrial level, and pre-combustion and oxy-fuel combustion lack successful industrial-scale demonstrations.

Table 12: Summary of the principal differences between the main types of carbon capture technology

Carbon Capture Technology	Advantage	Disadvantage	Reference
Post-combustion	Retrofitted to plants easily. Very developed technology with numerous full-scale commercial plants.	Dilute CO_2 concentration causing low driving force. Significant energy penalty of amine scrubbing process. Extensive water consumption	(Figueroa et al., 2008), (Mondal, Balsora and Varshney, 2012), (Wang et al., 2017)

Pre-combustion	■ Well established. ■ High CO_2 concentration and pressure.	■ Solvent or cryogenic separation required (energy penalty due to regeneration and more issues). ■ Low availability and poor operational flexibility of gasification technology. ■ High investment cost.	(Figueroa et al., 2008), (Mondal, Balsora and Varshney, 2012), (Wang & Song, 2020)
Oxy-fuel combustion	■ Very high CO_2 concentration. ■ No solvent required; can separate CO_2 with condensation.	■ High capital costs due to air separation technology. ■ ASU unit results in efficiency drop. ■ Lack of large-scale industrial demonstrations	(Basile et al., 2011), (Croiset and Thambimuthu, 2001), (Mondal, Balsora and Varshney, 2012), (Scheffknecht et al., 2011), (Wang & Song, 2020)

Using an IECM modelling software, a study by Rosa et al. (2021) compared the water consumption per tonne of CO_2 absorbed for each carbon capture method, the results of which are presented in Table 13.

Table 13: Simulated water consumption for each carbon capture method *(data retrieved from Rosa et al., 2021)*

Capture method	Water consumption (m^3/tonne CO_2) (presented as *median [low percentile; upper percentile])*
Post-combustion	1.71 [0.50; 2.33]
IGCC (Pre-combustion)	0.74 [0.65; 0.80]
Oxy-fuel combustion	2.22 [1.93; 2.69]

The results indicate that pre-combustion is much more water efficient than the other two options, which show similar results of water consumption.

5.2.1. Life Cycle Assessments

A life cycle assessment (LCA) is imperative to understand the environmental impacts of the carbon capture methods. The impact categories are the primary energy demand and the potentials for global warming (GWP), human toxicity (HTP), acidification (AP), photooxidant formation (POCP) and eutrophication (EP) (Schreiber, Zapp and Kuckshinrichs, 2009; Sifat and Haseli, 2019). Most important in this context is the GWP, which is a measure of the energy that one tonne of gas will absorb over a given period of time, relative to one tonne of CO_2, allowing the comparisons of the global warming impact of different gases (U.S. Environmental Protection Agency, 2023). In the context of carbon capture, GWP is useful to see how much CO_2 emissions are captured by the carbon capture methods. A summary of various LCAs are shown in Table 14.

Table 14: Summary of life cycle assessments undergone on carbon capture technologies

Technology Assessed	Conclusions	Reference
Post-combustion (MEA)	■ GWP for a plant with post-combustion carbon capture is much lower than a plant without. ■ HTP and EP are much higher (up to three times) with post-combustion carbon capture.	(Schreiber, Zapp and Kuckshinrichs, 2009)
Post-combustion, pre-combustion and oxy-fuel	■ Carbon capture plants emit more per kWh than assumed in clean-coal concepts (total CO_2 reduction by 72-90% and total greenhouse gas reduction by 79%) and substantially more when compared to renewable electricity. ■ However, carbon capture retrofit technologies would still greatly decrease expected CO_2 emissions.	(Viebahn et al., 2007)
Post-combustion, pre-combustion and oxy-fuel	■ Post-combustion carbon capture would result in increases in all categories, except GWP and acidification, due to solvent degradation and energy penalty. ■ Pre-combustion results in decreased impact in all categories (compared to post-combustion) due to efficient IGCC technology. ■ Oxy-fuel's effects are not known until energy demand for air separation is lowered and feasibility of co-capture of pollutants other than CO_2 is addressed (SO_x, NO_x etc.).	(Pehnt and Henkel, 2009)

Cuéllar-Franca and Azapagic (2015) compiled many studies to graph the GWP of various power plants with no capture, and also the three carbon capture methods. The results are shown in Figure 18.
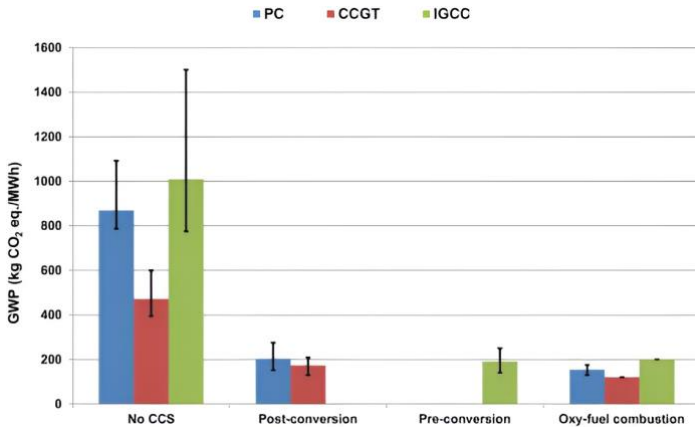

Figure 18: Global warming potential of CCS options for PC, CCGT and IGCC plants. PC: pulverised coal; CCGT: combined cycle gas turbine; IGCC: integrated coal gasification combined cycle (image retrieved from Cuéllar-Franca and Azapagic, 2015).

This analysis indicates that post-combustion and pre-combustion (referred to as post-conversion and pre-conversion in Figure 18) possess similar GWP with oxy-fuel combustion displaying a much lower GWP if pulverised coal and a combined cycle gas turbine are used. This suggests that the energy penalty of gasification greatly lowers the theoretical efficiency of IGCC technology.

The LCAs indicate that the GWP is significantly reduced by any carbon capture method. However, there is ongoing debate on whether the environmental consequences of solvent regeneration, degradation and toxicity are more damaging than the energy penalty of gasification. Oxy-fuel combustion is generally seen as the most environmentally friendly option, but further research is needed to validate this claim.

5.2.2. Techno-Economic Analyses

In addition to Life Cycle Assessments, it is imperative to analyse techno-economic analyses to assess the technologies. The best method of assessing the cost of carbon capture technology

is calculating the levelized cost of electricity (LCOE) and then the CO_2 avoided cost (AC). The levelised cost of electricity (LCOE) is the constant dollar electricity price that would be required over the life of the plant to cover all costs. The CO_2 avoided cost is the cost to remove a tonne of CO_2. The methods for calculating are shown by Hanak et al. (2017):

$$LCOE = \frac{TCR \times FCF \times FOM}{\dot{W}_{net} \times CF \times 8760} + VOM + \frac{SCF}{\eta_{th}} \quad (11)$$

$$\frac{SCF}{\eta_{th}} = \frac{\dot{W}_{net}}{\dot{Q}_{fuel}} \quad (12)$$

$$e_{CO_2} = \frac{\dot{m}_{CO_2}}{\dot{W}_{net}} \quad (13)$$

$$AC = \frac{LCOE_{capture} - LCOE_{ref}}{e_{CO_2,ref} - e_{CO_2,capture}} \quad (14)$$

The equations include thermodynamic performance indicators such as net power output ($\dot{W}_{net}$), net thermal efficiency (η_{th}), heat input from fuel combustion ($\dot{Q}_{fuel}$), capacity factor (CF) and specific CO_2 emissions (e_{CO_2}). They also include economic performance indicators such as total capital requirement (TCR), variable (VOM) and fixed (FOM) operating and maintenance costs, specific fuel cost (SFC), and the fixed charge factor (FCF), which considers the system's lifetime and project interest rate.

A study undergone by the International Energy Agency Greenhouse Gas Programme (IEA GHG) (2006) assessed the performance and costs of various power stations incorporating carbon capture. The study approached 100 public and private power utilities, with 34 organisations from 17 countries responding. The cost of CO_2 compression to 11MPa for pipeline transport was included in the cost of electricity generation. The costs of CO_2 transport and storage were excluded as these depend significantly on location conditions. The results are shown in Table 15.

Table 15: Comparison of coal fired, and gas fired power stations with or without CO_2 capture
(retrieved from International Energy Agency Greenhouse Gas Programme, 2006)

Technology	Thermal efficiency [%LHV][a]	Capital cost [$/kW]	Electricity cost [c/kWh]	Cost of CO_2 avoided (AC) [$/tonne CO_2]
Coal Fired Plants				

No capture	44.0	1410	5.4	-
Post-combustion capture	34.8	1980	7.5	34
Pre-combustion capture	31.5	1820	6.9	27
Oxy-fuel combustion	35.4	2210	7.8	36
Gas Fired Plants				
No capture	55.6	500	6.2	-
Post-combustion capture	47.4	870	8.0	58
Pre-combustion capture	41.5	1180	9.7	112
Oxy-fuel combustion	44.7	1530	10.0	102

a – LHV refers to Low Heating Value, which is an thermal efficiency that does not include the condensation of water vapour produced by the combustion process (Fulton, 2021).

This study shows the wide difference between the costs and thermal efficiencies of carbon capture for coal and gas fired power plants. Gas fired plants are shown to have much higher thermal efficiency and lower costs. For gas-fired plants, post-combustion capture was shown to have the highest thermal efficiency and lowest cost of CO_2 avoided. The discrepancy is likely due to the high energy demands for gasification and oxygen production for pre-combustion and oxy-fuel combustion respectively. Among coal-fired plants, it was determined that pre-combustion capture is the most efficient and economically viable method. Oxy-fuel was shown to be the least efficient for both but pre-combustion had a higher AC for gas-fired plants. These findings highlights the need for adapting the method based on the specific conditions of each power plant.

A study undergone by Massachusetts Institute of Technology (MIT) (2007) also compared the three technologies, using 500W net output as a basis and using coal as the fuel, the results of which are shown in Table 16. Like in the previous study, transportation and storage costs were not included.

Table 16: Representative performance and economics data for the three main capture technologies *(using data from Massachusetts Institute of Technology, 2007).*

Technology	Generating Efficiency [%][a]	Capital Cost ($/kWe)	LCOE (c/kWh)	Cost of CO_2 avoided ($/tonne CO_2)
No capture	38.5	1330	4.78	-
Post-combustion capture[b]	29.3	2140	7.69	40.4

| Pre-combustion capture | 31.2 | 1900 | 6.98 | 24.0 |
| Oxy-fuel combustion | 30.6 | 1890 | 6.52 | 30.3 |

a – Generating efficiency refers to the proportion of thermal energy in the fuel derived from the fuel that ultimately contributes to the net electricity produced.
b - Post-combustion capture was performed with a MEA amine solution.

In this study, pre-combustion has the lowest cost of CO_2 avoided, which coincides with the conclusions of the IEA GHG study for coal-fired plants. However, in this study, post-combustion capture has the highest cost of CO_2 avoided rather than oxy-fuel, revealing a lack of consensus.

The efficiency penalties varied for each technology: for post-combustion, the largest efficiencies were for the CO_2 recovery (-5.7%) and CO_2 compression (-3.5%). For pre-combustion carbon capture, the efficiency losses for CO_2 compression and recovery were much lower (at -2.1% and -0.9%, respectively) due to the higher pressure and higher CO_2 concentration of the flue gas. Despite this, there was a high penalty for the water shift process (-4.2%). Oxy-fuel combustion showed a high penalty for CO_2 compression (-3.5%) but the highest penalties were for the ASU (-6.4%) and the boiler/FGD (-3%). It is important to note the lower compression penalty in the pre-combustion case as compared to the other two. The study indicates that pre-combustion is the most efficient carbon capture technology. However, oxy-fuel combustion could be the most efficient if the energy penalty for the ASU were lowered.

The comparisons reveal a lack of consensus on the most efficient carbon capture technology. However, the studies suggest that the energy penalties associated with the ASU and gasification units are the most significant for pre-combustion and oxy-fuel, respectively. Many of the studies indicate pre-combustion to be the most efficient option in terms of water efficiency, performance in the LCA and efficiency for coal fired plants. The conclusions for oxy-fuel combustion varied widely, for instance it was shown to be the least efficient for both fuels in the simulation by the International Energy Agency Greenhouse Gas Programme but had a good generating efficiency and AC in the MIT study. Its fluctuating performance is most likely due to its novelty as a technology and an industrial-level demonstration is required to truly know its performance.

The LCAs and techno-economic analyses indicate that post-combustion carbon capture has many problems that must be addressed by newer technologies. Amine-based post-combustion capture is a well-developed technology which failed the LCA and thus development should be directed towards pre-combustion and oxy-fuel combustion carbon capture, which show much more promise.

6. DISCUSSION

In this section, the interpretation and implications of the findings derived from our theoretical research study on carbon capture are explored. The aim of this project was to provide a clear perspective on the current state of carbon capture technology and to assess its effectiveness on mitigating the effects of climate change.

The findings indicate that post-combustion carbon capture using MEA solvent is the most developed carbon capture technology. Contrary to initial expectations, this method can have a negative effect on the environment due to the toxicity of MEA and its degradation products. This was reflected in the poor results of post-combustion carbon capture in the life cycle assessment. The literature review revealed that there are many alternatives to MEA as the solvent, however the majority of these have only been tested at a pilot scale and must be scaled up to be seen as legitimate alternatives.

The research also highlighted pre-combustion and oxy-fuel combustion carbon capture as very promising technologies. Overall, pre-combustion can be seen as the most efficient technology from current studies (this can change depending on the fuel utilised), however more demonstrations at large scales are required to conclude this. The studies highlighted the energy penalty of gasification, which should be the focus of pre-combustion research.

The findings on oxy-fuel combustion carbon capture expressed that it could be the most efficient carbon capture technology, however, like in pre-combustion, more studies are required. The ASU requirements were identified as the major disadvantage of oxy-fuel combustion. Therefore, if a sufficiently efficient and powerful ASU were to be developed, oxy-fuel would likely become the best option. More research into ASU is required and a comparison of the technologies in the future when ASUs are more developed is imperative.

For all the technologies, the efficiency drops indicate that more fuel will be needed to generate the same amount of energy. This offsets the environmental benefit of carbon capture. However, carbon capture is shown to be an overall positive as the GWP from the Life Cycle Assessment is much lower regardless of which capture method is adopted.

In addition, this efficiency drop would result in higher fuel costs, as indicated by the high CO_2 avoided cost associated with carbon capture. These drawbacks would undoubtedly lead to significant economic and societal effects, which are beyond the scope of this project but should be investigated. Furthermore, carbon capture will require some form of carbon credit system to incentivize its use, due to the inherent economic disbenefits to the process (for all types). This is imperative as carbon capture may be neglected to prioritise short term profits over climate change mitigation. In the future, climate change will bring about long-term effects which will render short-term profits irrelevant.

The investigation into transport and storage revealed substantial drawbacks to carbon capture. Primarily, the possibility of leakage could render storage of CO_2 useless on a long-term scale. This indicates that the field of CO_2 storage is the most crucial as emissions are only reduced if the captured CO_2 can be safely stored. The research also showed that the storage requirements are extensive and need continuous expansion. Future research should focus on CO_2 leakage prevention and expanding storage options.

Furthermore, enhanced oil recovery as the primary destination for captured CO_2 renders carbon capture very inefficient for climate change mitigation. This is due to the net emission increase resulting from the process when the recovered oil is combusted. Moreover, if carbon capture technology is used to continue to combust fossil fuels, then it could result in sustaining fossil fuel industries rather than mitigating climate change.

Hence, at the current state of carbon capture, taking into account the disbenefits of amine-based post-combustion carbon capture, the possibility of CO_2 leakage and the use of enhanced oil recovery, renewables are markedly more effective than carbon capture. This is because renewables do not emit CO_2 directly and do not produce waste in their operation. If carbon capture promotes the development of more fossil fuel-based power plants, they are most likely to be negative for climate change mitigation. This is further compounded due to the economic investments into carbon capture that could be invested into renewables, or other alternatives such as nuclear and energy efficiency methods. However, if pre-combustion and oxy-fuel are developed to become more viable and the majority of captured CO_2 would be stored in a leakage-free manner, then carbon capture would be a great option for climate change mitigation.

While this project provides valuable insights, it has many limitations. Firstly, many comparisons were conducted using simulations from other authors. Simulations are often flawed

as they fail to consider many factors that arise in industrial operation. Additionally, the simulations often used different parameters, reducing the ability to compare between them. Furthermore, sources were used from a variety of dates, with the vast majority being from this millennia. However, due to the fast-developing nature of carbon capture, even recent sources could have outdated information. This can only be avoided by newer comparisons of the technologies with similar parameters.

The sources for this study were mostly from environmental organisations and from peer-researched journals. Books were rarely used due to the novelty of carbon capture technology, rendering accessible books outdated and thus, irrelevant. This was particularly the case for alternatives to amines for post-combustion carbon capture and for the technologies of pre-combustion carbon capture and oxy-fuel combustion carbon capture.

The implications of the findings extend beyond theoretical considerations. The further development of climate change mitigation technologies, whether that be carbon capture or other technologies, is crucial to avoid its consequences. As stated in the introduction, climate change will affect the entire world. Further research must expand upon the findings presented to further assess the viability of carbon capture.

In conclusion, this theoretical analysis indicates that carbon capture is a promising technology but is currently not effective in mitigating climate change due to the energy penalties and environmental disbenefits of the technology in its current state. The destination of carbon capture has been identified as the primary concern. The analysis also highlights improved efficiency of gasification and ASU as the most imperative areas of research, to improve the pre-combustion and oxy-fuel combustion carbon capture technologies.

N. CONCLUSIONS

This project provides a comprehensive review of the currently available carbon capture technologies and assessed these technologies on their ability to mitigate climate change. Through a comprehensive analysis of the literature, several key findings have emerged.

Firstly, an amine-based post-combustion carbon capture process, particularly using MEA, was shown to be the most developed technology. Despite this, many problems are associated with an amine-based process, including corrosion, high solvent regeneration cost, water consumption, solvent degradation, and toxicity. A literature review revealed that much research is directed into alternatives to amine solvents, including zeolites, MOFs and membranes. Nevertheless, these alternatives are currently in the early-stage development and more research is required to assess their viability.

Pre-combustion carbon capture was shown to be an effective alternative, as the resulting flue gas has a much higher pressure and higher concentration of CO_2 compared to post-combustion due to gasification technology. This results in theoretically higher efficiency, which was supported by techno-economic analyses (however this was not confirmed definitively). Physical absorption processes were shown to be viable (with research indicating that Selexol was the most viable) and cryogenic separation was shown to be promising but energy intensive.

Oxy-fuel combustion was shown to be potentially the most efficient technology due to the very high concentration of CO_2 in the flue gas resulting from the absence of nitrogen. However, the findings highlighted that oxy-fuel combustion has a very high oxygen requirement, and the ASU remains the greatest limitation in terms of cost and energy penalty. Furthermore, the technology was shown to be very new, and many studies could not confirm its viability because of this.

Techno-economic analyses revealed high energy penalties for all the carbon capture technologies and as a result, high costs of CO_2 avoided were calculated. The efficiencies and costs of CO_2 avoided changed dramatically depending on the study and the fuel used. Life Cycle Assessments showed that all carbon capture technologies reduced the Global Warming Potential,

however an amine-based post-carbon capture performed poorly primarily due to the toxicity of MEA and its degradation products.

Research into the transport of CO_2 revealed that pipelines is the most common form of transport and that the pressurisation and transport of CO_2 inherits a high energy penalty, which is lowest for pre-combustion carbon capture.

Storage was shown to be a key challenge for carbon capture. The storage requirements to achieve climate change mitigation goals were established as high. Crucially, the leakage of CO2 from geological sequestration was identified as a paramount issue and a gap in current research as the leakage rate is not known.

Enhanced oil recovery was revealed to be a primary destination for captured CO_2, which was demonstrated to offset the benefits of carbon capture as it most likely results in positive net emissions for the carbon capture process despite its economic benefits.

In the discussion, the current state of carbon capture was argued to be poor to mitigate climate change due to the issues associated with post-combustion carbon capture in addition to the possible leakage of CO_2 and the use of CO_2 in enhanced oil recovery. Renewables were put forth as a better alternative in the current state, but it was also argued that this could change with development of pre-combustion and oxy-fuel carbon combustion capture technologies. Political decisions were advocated to promote the use of carbon capture and storage, including carbon tax credits and a focus on geological storage of captured CO_2 over enhanced oil recovery.

In conclusion, this project has highlighted pre-combustion and oxy-fuel combustion carbon capture as the most promising carbon capture due to the challenges associated with post-combustion carbon capture. At its current state, carbon capture was shown to be inefficient for climate change mitigation goals but, optimistically, further research and development into carbon capture will resolve its issues and increase its efficiency, whilst also assuring the permanent storage of capture carbon dioxide.

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ACRONYMS

AP – Acidification Potential

CCS – Carbon Capture and Storage

CF – Capacity Factor

CMS – Carbon Molecular Sieve

DCC – Direct Contact Cooler

DGA – Di-glycol amine

DEA – Di-ethanol amine

EOR – Enhanced Oil Recovery

EP – Europhication Potential

FCF – Fixed Charge Factor

FGD – Flue Gas Desulphurisation Unit

FOM – Fixed Operating and Maintenance Costs

GWP – Global Warming Potential

HTP – Human Toxicity Potential

IGCC – Integrated Gasification Combined Cycle

IEA GHG - International Energy Agency Greenhouse Gas Programme

LCA – Life Cycle Assessment

LHV – Low Heating Value

MDEA – Methyl-diethanol amine

MEA – Mono-ethanol amine

MOF – Metal Organic Framework

NETL – National Energy Technology Laboratory

NG – Natural Gas

PC – Pulverised Coal

PSA – Pressure Swing Adsorption

SFC – Specific Fuel Costs

TCR – Total Capital Requirement

TSA – Temperature Swing Adsorption

UNECE – United Nations Economic Commission for Europe

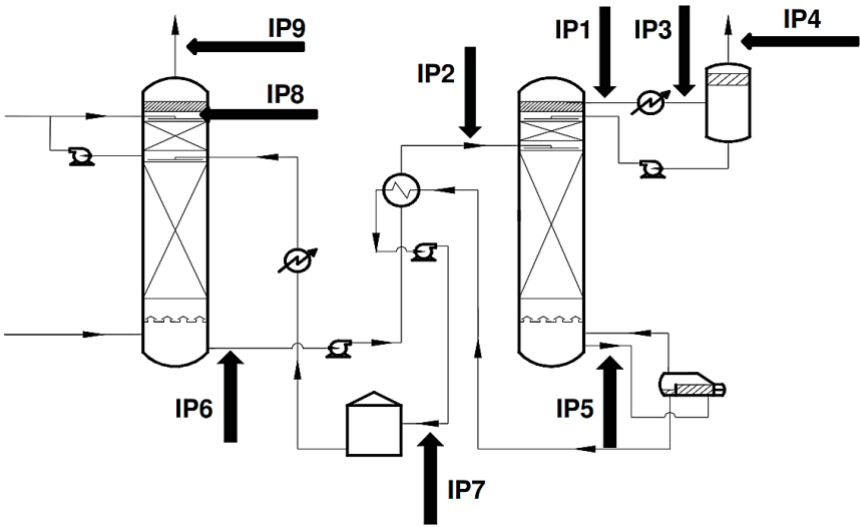VOM – Variable Operating and Maintenance Costs

APPENDICES

APPENDIX

Figure I: Schematic CO2 capture unit and corrosion monitoring insertion points *(image retrieved from Kittel et al., 2009).*
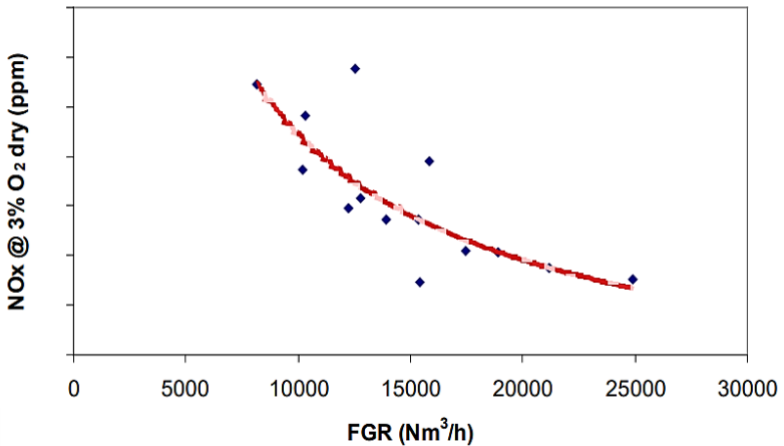
Figure II**:** Relationship between NOx emissions and FGR rate for Total oxy-fuel pilot plant
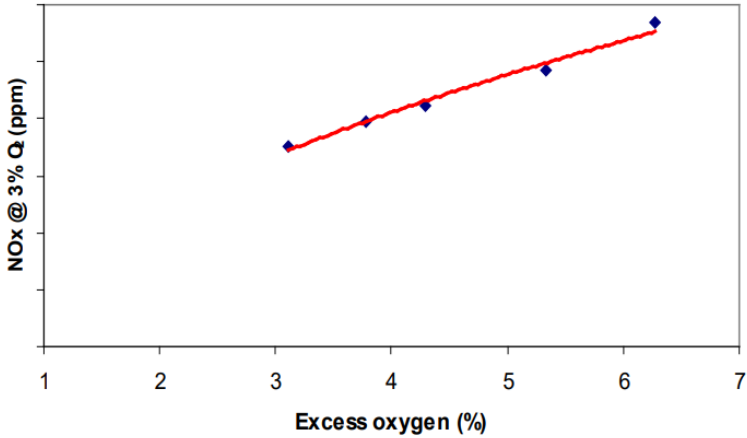*(image retrieved from Marcano et al., 2011).*

Figure III: Relationship between NOx emissions and excess oxygen for Total oxy-fuel pilot plant
pilot plant *(image retrieved from Marcano et al., 2011).*