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**Environmental Assessment of Post-Combustion Carbon Capture Process Retrofitting
in Power Plant: Life Cycle Assessment**

**A Thesis Submitted in Partial Fulfilment of the Requirements for
the Doctor of Philosophy Degree in Environment and Sustainable
Development**

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Abstract

Carbon dioxide (CO₂) emission from burning fossil fuels in power plants is a major contributor toward climate change. Carbon capture and storage (CCS) is a potential solution to reduce the emission while renewable energy sources are being developed. Among CCS technologies, post-combustion carbon capture (PCC) is a promising method for reducing CO₂ from existing power plants. However, PCC has drawbacks, including high construction costs, high energy demand for solvent regeneration, and potential environmental impacts.

A stepwise multi-linear regression based on the least squares method was used to identify significant variables in PCC to reduce solvent regeneration energy and to enhance efficiency. Seven independent variables were selected, including lean solvent temperature, solvent concentration, lean CO₂ loading, and flue gas conditions. The analysis revealed that solvent concentration, inlet CO₂ concentration, and lean CO₂ loading affect solvent regeneration energy. The regression equations were validated and found reliable, with prediction errors of 0.78% for CO₂ rich loading and 2.47% for solvent regeneration energy.

A study-state rate-based model of PCC system using monoethanolamine (MEA) as a solvent was developed and validated in Aspen HYSYS using pilot plant data. The model predictions using HYSYS agreed with PCC pilot plant experimental data where the deviation between the model and experimental results did not exceed $\pm 8\%$. A new simulation of PCC process using combined solvent MDEA/PZ under different total concentration (40 wt% and 50 wt%) and different individual concentration for MDEA and PZ was simulated. The aim of the simulated PCC process using combined solvents was to investigate the synergistic effect of the selected solvents compared to MEA at 30 wt%. The simulation shows that 43 wt% MDEA and 7 wt% PZ is the optimum composition that requires the minimum energy for solvent regeneration of 2.53 MJ/kg_{CO2} for 88.5% process efficiency compared to 4.003 MJ/kg_{CO2} for 30 wt% MEA without the need for any process modifications.

A cradle-to-gate life cycle assessment is used to investigate and evaluate the environmental impact of PCC once retrofitted to power plant using SimaPro. Three methods of life cycle assessment were used in this study including ReCiPe 2016 Midpoint (H), ReCiPe 2016 Endpoint (H), and IPCC 2013 GWP 500. The findings from LCA study show that power plant equipped with PCC demonstrated a tremendous reduction of global warming potential compared to power plant without PCC. However, the study found that power plants with or without PCC cause resource depletion due to the utilization of fossil fuel. Furthermore, several

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environmental issues impact specifically water resources reach the maximum once MEA is utilized for PCC compared with MDEA/PZ that required of water management system to manage the water utilization and damping during PCC process. Hence, the study shows the synergistic effect of MDEA and PZ to capture carbon dioxide using PCC process shows the ability to resolve the drawback of PCC process from technical and environmental view and could be selected as one of the mitigation solutions to reduce CO₂ emissions. However, the study limited to the economic assessment for the system and the variation in the power plant process as well as the transportation and storage stages of carbon capture and utilization or storage process.

Publications and Presentations

- **Alalaiwat, D.**, and Khan, E. (2023) Current status and future scenarios of carbon capture from power plant emissions: a review. *Reviews in Environmental Science and Bio/technology*. 22, 799-822.
- Bahman, N., **Alalaiwat, D.**, Abdulmohsen, Z., Al Khalifa, M., Al Baharna, S., Al-Mannai, M., Younis, A. (2023) A critical review on global CO₂ emission: where do industries stand? *Reviews on Environmental Health*. 38 (4), 681-696.
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Dedication

I dedicate this PhD thesis to *my family*, whose unwavering support and love have been my constant source of strength.

To my supervisor Dr. Ezzat Khan, thank you for your guidance, support, and belief in me.

To my friends, for being in my life and providing me with encouragement in every moment of my study.

To all those who have dreams, strive, and persevere in the pursuit of knowledge.

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List of Abbreviations

ADP	Abiotic Depletion Potential
BP	British Petroleum
CSIRO	Commonwealth Scientific and Industrial Research Organization
CCUS	Carbon Capture Utilization and Storage
CCS	Carbon Capture and Storage
CC	Carbon Capture
CCU	Carbon Capture and Utilization
CCGT	Combined Cycle Gas Turbine
DAC	Direct Air Capture
ENRTL	Electrolyte Non-Random Two-Liquid
EP	Eutrophication Potential
EU	Energy Use
FD	Fossil Depletion
FROLS	Orthogonal Least Squares
FWEP	Freshwater Ecotoxicity Potential
GWP	Global Warming Potential
GHG	Greenhouse Gas
HTP	Human Toxicity Potential
HSS	Heat Stable Salts
IEA	International Energy Agency
IPCC	The Intergovernmental Panel on Climate Change
ISO	International Organisation for Standardisation
LCA	Life Cycle Assessment
MAE	Marine Aquatic Ecotoxicity
MEA	Monoethanolamine
MDEA	Methyldiethanolamine
MDP	Metal Depletion Potential
MEP	Marine Ecotoxicity Potential
NARX	Polynomial Nonlinear Auto-Regressive with Exogenous
OCGT	Open Cycle Gas turbine
ODP	Ozone Depletion Potential
OFTEP	Ozone Formation Terrestrial Ecosystem Potential
OFHHP	Ozone Formation Human Health Potential
PC	Pre-Combustion
PCC	Post-Combustion Capture
PHOF	Photochemical Oxidant Formation
PMF	Particulate Matter Formation
PSA	Pressure Swing Adsorption
PI	Process Intensification
PZ	Piperazine

List of Abbreviations

SDG	Sustainable Development Goals
SPSS	Statistical package for Social Science 25
TAP	Terrestrial Acidification Potential
TEP	Terrestrial Ecotoxicity Potential
TSA	Temperature Swing Adsorption
UN	United Nations
VIF	Variance Inflation Factor
VSA	Vacuum Swing Adsorption
WCP	Water Consumption Potential
WU	Water Use

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Chapter 1

Introduction

Introduction

1.1 Overview

This chapter sets the content of the thesis. Section 1.2 presents the background of the study that is divided into three subsections: energy demand and CO₂ emissions that affect climate change, power plant challenges, and the CO₂ capture options. The aim and objectives of this thesis are presented in Section 1.3 while the problem statement is presented in Section 1.4. In Section 1.5 research questions are listed. The link of this study to the Sustainable Development Goals (SDG) is presented in Section 1.6. The significance of the study is outlined in Section 1.7. In Section 1.8 the limitations of the study are presented. The structure of the thesis is presented in Section 1.9 and the definition of the terms belonging to the study and being used in the following text is outlined in Section 1.10.

1.2 Background

1.2.1 Energy demand, CO₂ emissions and climate change

The economic growth, urbanization, transportation beside the population growth is expected to grow about 1.7 billion by 2050 are the drivers of the global energy demand (IEA, 2023). The global energy demand grew by 0.7% in 2023 compared to 0.5% in 2019 (IEA, 2019, 2023). Furthermore, the British Petroleum (BP) report shows that the global energy demand increased by 6% in 2021 compared to 2020 (BP, 2022) specifically after the locked down period during COVID-19 pandemic. Power plants are the main energy providers that generate electricity using fossil fuels (natural gas or coal) (Chao et al., 2021). The demand on coal and natural gas for power plant exceeds the other sectors that reached 54% for coal and 39% of natural gas. (IEA, 2023).

Growing the utilization of fossil fuel for energy generation since the mid of 20th century has increased the emissions of Greenhouse Gases (GHG) (Oh et al., 2016). Among the GHGs, CO₂ is the most contributor to the climate change and global warming (IPCC, 2005). The combustion of fossil fuels increased the emissions of CO₂ from energy production, industrial processes, flaring and methane by 5.9% in 2021 compared to 2019 (BP, 2022). It is also considered the largest stationary source of CO₂ emissions (IEA, 2017), although the utilization of renewable energy increases but yet unable to replace power plant processes using fossil fuels (Sultan et al., 2021). The energy outlook of International Energy Agency

(IEA) stated that as far as fossil fuel is the solely energy provider, the energy demand and CO₂ emissions will keep rising (IEA, 2018).

The GHGs emission has contributed to increase atmospheric CO₂ reaching 413.2 ppm in 2020 compared to 361.7 ppm during the period of 1991-2000, this increase is responsible for about 81% of the increase of radiative forcing (IPCC, 2023). It resulted in global warming (Rehman et al., 2023), that causes to rise atmospheric temperature and the ultimate change in climate (Akinola et al., 2019). The Intergovernmental Panel on Climate Change (IPCC) issued a special report (SR15), addressing the critical consequences of global warming if it increases by 2°C compared to the preindustrial level. The IPCC report shows that the risk from precipitation, drought, rise of sea level and the damage to many ecosystems will be higher at 2°C compared to 1.5°C. Therefore, IPCC recommended solutions to reduce CO₂ emissions by improving electrification, increasing the contribution of alternative energy sources such as hydrogen and bio-fuels and investing more in Carbon Capture Utilization and Storage (CCUS) (IPCC, 2018). According to IEA report, 1Gt_{CO2} per year must be captured from power, iron, cement, and chemical plants by 2030 to enable achievement of net zero emissions (IEA, 2020). Research and development related to Carbon Capture and Storage (CCS) technology and CCUS have increased over the past decade as one of the promising methods to reduce CO₂ emissions, specifically from power plants (Oh et al., 2018; Tiwari et al., 2023). Among various CO₂ emissions reduction methods, CCS is expected to contribute by approximately 19% reduction in the total CO₂ emissions by 2050 (IEA, 2011).

1.2.2 Power plant function and challenge

The thermal power plant generates electricity through combustion of fossil fuels, either coal or natural gas, to produce steam that drives the turbine using Rankine cycle since 1882 (Veselov et al., 2019). Power plants generating electricity using natural gas are divided into two types: “Open Cycle Gas turbine” (OCGT) and “Combined Cycle Gas Turbine” (CCGT). The working principles of OCGT are based on Brayton cycle whereas the power is generated using gas turbine through a shaft, the efficiency of the power production is within 35-42% based on lower heating value. The CCGT produces extra power using a heat recovery steam generator to recover heat from exhaust gases thereby improving thermal efficiency up to 60%. Over the last few decades, the CCGT technology is utilized more compared with OCGT due to higher power production and better efficiency

(Rao & Francuz, 2013). The power plant generating electricity using coal is the coal-fired power plants that are classified as pulverized coal plant, fluidized bed combustion, and integrated gasification combined cycle (Romero-García et al., 2022). However, coal-fired power plants contain complex system to burn coal effectively and to cool the heat generated during the process (Mac Dowell & Shah, 2014).

The operation of power plants face several challenges including fuel pricing, environmental concerns, fossil fuel depletion, regulations and transition to renewable energy (IEA, 2015). These challenges compel the energy providers to introduce technological innovations and introduce methods to control the negative effects of the power plant operation on the environment (Fu et al., 2021; Veselov et al., 2019). Therefore, exploring new technologies to increase the thermal efficiency that raises the steam temperature and reduces gas emission is the main concern through out the globe (Xu et al., 2018).

The emissions of CO₂ from power plants, using coal and natural gas, reached their highest ever level of 46% of the global increase in emissions where coal ranked the first by 40% among the others fossil fuels (IEA, 2021c).

1.2.3 CO₂ capture options

Primarily, Carbon Capture (CC) stage is the most expensive stage of CCS process, 50-90% of the overall cost of the process is taken by captured stage (Pera-Titus, 2014). There are several variables need to be considered prior to building a CC system for instance, the amount and type of solvent or sorbent utilized, the pre-treatment requirements of impurities, environmental impact of the chemicals, the concentration of CO₂ in the flue gas, and the selected carbon capture techniques (Gür, 2022). Three techniques are currently available for CC from stationary system, the pre-combustion CC, oxy-combustion capture, and post-combustion capture. The advantages and disadvantages of the three technologies are summarized in **Figure 1**, among these the post-combustion capture is extensively used and attracted the researcher's attention (Bui et al., 2018; Kearns et al., 2021; Pan et al., 2016). Furthermore, post-combustion capture is the selected technique for this study.

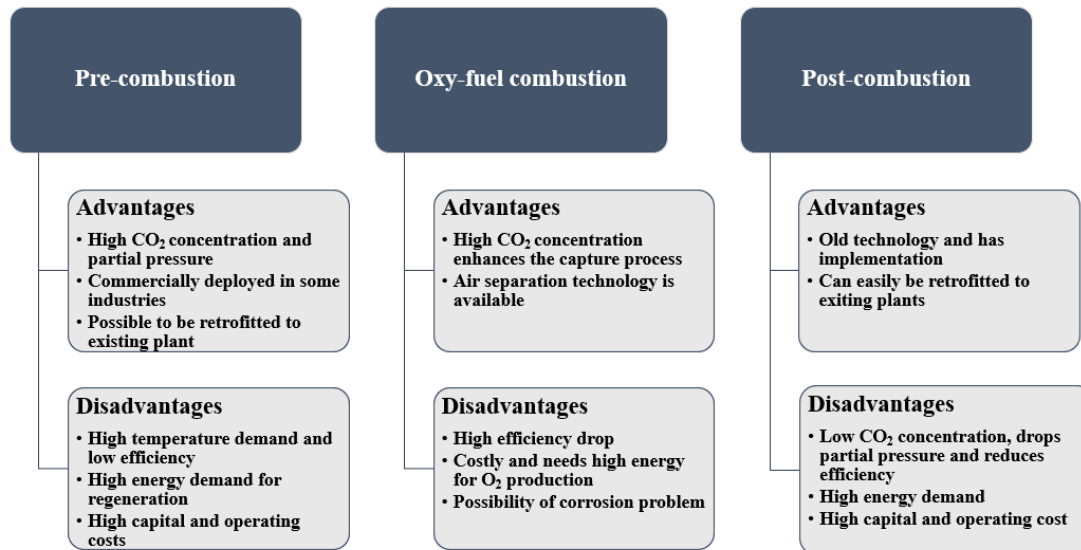


Figure 1 Advantages and disadvantages of the carbon capture technologies including (1) Pre-combustion capture, CO₂ is captured from fuel before burning using air, (2) Oxy-fuel combustion capture, CO₂ is captured from flue gases after burning fossil fuel using pure oxygen, and (3) Post-combustion capture, CO₂ is captured from flue gases after burning with air.

1.2.3.1 Pre-combustion carbon capture (PCC)

In this method, the CO₂ is captured before burning the fuel in the combustion chamber (Eide & Bailey, 2005). The fossil fuel such as coal is reacted with steam and oxygen at elevated temperature and pressure that results in synthesis gas, contains CO and H₂ (Cormos, 2012; Jansen et al., 2015). Afterwards the CO is converted to CO₂ using steam therefrom CO₂ is captured outside the stream using solvent such as Selexol, the pure hydrogen gas is burned to generate electricity (Rubin et al., 2012) as shown in **Figure 2**. The produced CO₂ can be captured from natural gas by a reforming process which is similar to the Pre-Combustion (PC) using coal that is used for hydrogen production (Rubin et al., 2012). Although the PC process can be operated in power plant for generating electricity using natural gas at lower energy than post-combustion capture, but the process is commercially less viable than post-combustion capture (Rubin et al., 2015). The pre-combustion CC is also used for hydrogen production including CO₂ capture unit known as blue hydrogen production (Rubin et al., 2012). The technique has gained more attraction in research and development, owing to the reason that it works toward climate change mitigation. The process could use coal or natural gas as a source of fuel where gasifier is used for burning the fossil fuel, followed by sending the

burning products for a shift reactor to ensure burning and formation of hydrogen (Rubin et al., 2012).

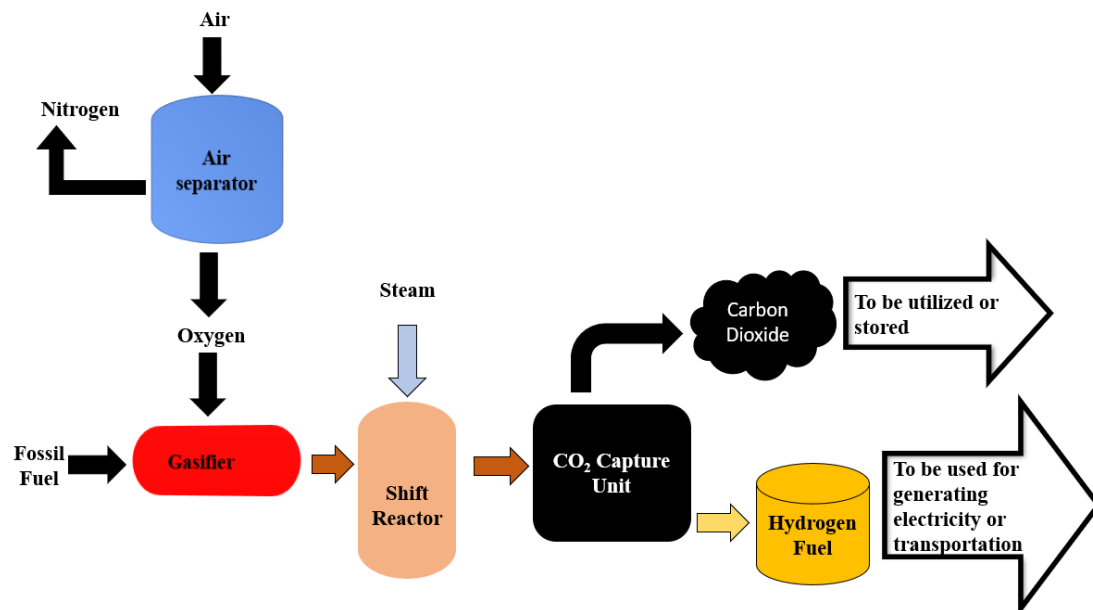


Figure 2 Schematic representation of pre-combustion CC process in power plants using coal as a source of fuel.

The driving force for the PC process is the solubility of CO₂ in the solvent being used for the purpose, that increases at high pressure and low temperature (Smith et al., 2022). The CC using PC process by solvent absorption techniques does not require chemical reaction due to the high CO₂ partial pressure. The solvents used are amines, amino acid salts, carbonate compounds, aqueous ammonia, immiscible liquids, ionic liquids, hydrophobic polymers and some other solvents that are able to reduce overall cost of the capture process, improve the absorption rate, reduce the solvent degradation, minimize the equipment size, and reduce the solvent regeneration energy (Mumford et al., 2015). The selected solvent supposedly has a high potential on the cost of the CCS process where the solvent properties have a direct effect on the economic cost of the process (Siefert et al., 2016).

1.2.3.2 Oxy-fuel combustion CC

Oxy-fuel combustion is an alternative proposed technique against the post-combustion capture process. The process is specifically used for coal-fired power plants where pure oxygen (95-99%) is used for burning instead of air at high temperature in the combustion chamber (Metz et al., 2005). The burning processes produce CO₂ that is separated without further processing

due to the high CO₂ concentration being formed (Tan et al., 2006). The CO₂ from the oxy-fuel combustion has 90% purity, is passed to be compressed and stored (Shaw & Mukherjee, 2022). The description of the oxy-fuel combustion CC is shown in **Figure 3**. It includes three stages, O₂ is separated from the air using air separation unit, pure O₂ is used to burn the fossil fuel and energy is generated, and finally the CO₂ formed during the burning processes is compressed and transported for storage.

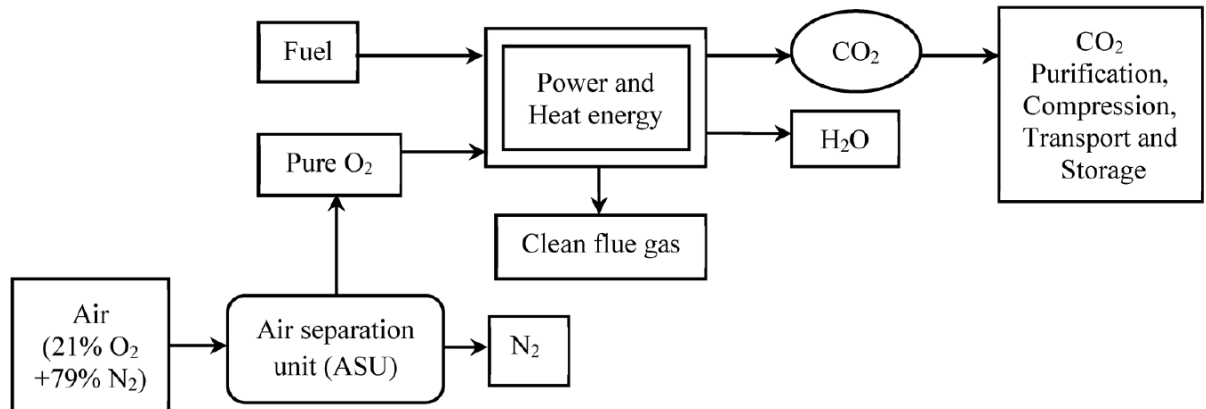


Figure 3 Description of oxy-fuel combustion CC process (Maniarasu et al., 2021).

The air separator unit is required for separating oxygen from air to be used for burning, is considered to be expensive (Rubin et al., 2012; F. Wu et al., 2018). The minimum energy required to separate oxygen from air is 176 MJ/t_{O₂} using air separation unit that is considered as a main process challenge (Fu & Gundersen, 2012). However, a new technique for separating of O₂ is introduced using membrane or oxygen capturing solids, that targets the reduction of energy demand (Zhang, 2015). Challenges such as high temperature and the risk of air leaks into the system (Yadav & Mondal, 2022) are still to be address, hence, oxy-fuel process is not expected to be of economic attraction unless more legislation for carbon price is issued (Wall et al., 2011).

1.2.3.3 Post-combustion carbon capture (PCC)

The post-Combustion Capture (PCC) has gained most of the attention and has been demonstrated in industries at different level, for example, TMC Mongstad in Norway that captures 300,000 tones/year of CO₂ and BD3 SaskPower in Canada captures 1 million tones/year, are the biggest PCC projects so far (Liang et al., 2015). In PCC process, the CO₂ is removed from the flue gas once fossil fuel is burnt (Hong, 2022) followed by storage of the captured CO₂, **Figure 4**. The PCC is applicable to capture CO₂ from

this sector to support the achievement of net zero emission (Gür, 2022). Once the flue gas is burnt to release energy in the form of heat, byproducts such as CO_2 , water vapor, SO_2 and Nitrogen Oxides (NO_x) are formed which require to be removed prior to CO_2 capture (Merkel et al., 2010). Currently, the PCC is grouped in to three categories; biological, physical and chemical methods (Maniarasu et al., 2021).

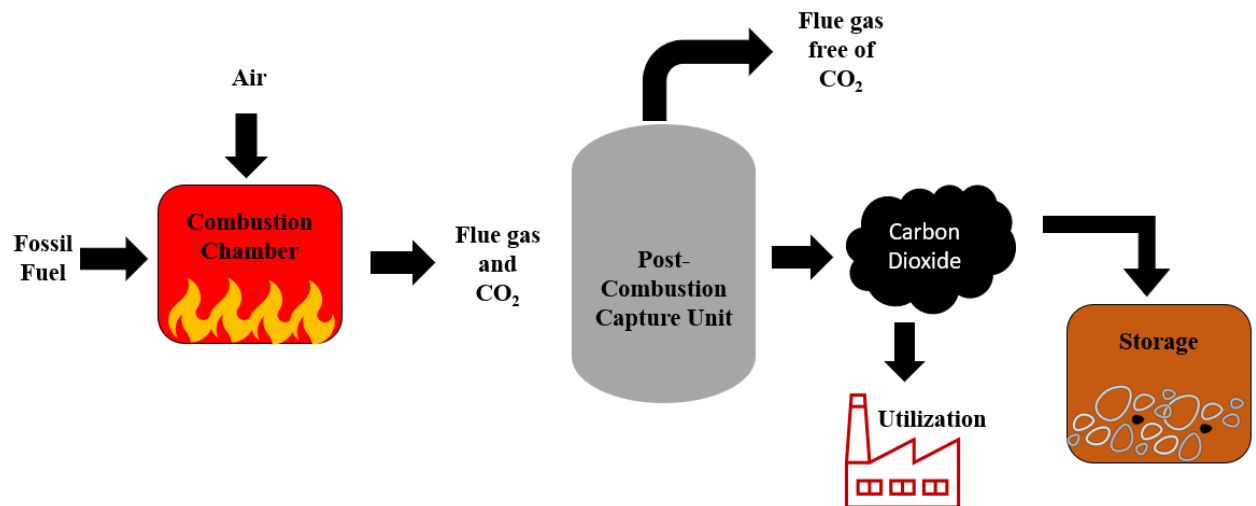


Figure 4 Process block diagram of PCC. The flue gas contains CO_2 leaves the combustion chamber passed through the capture system. The clean flue gases and CO_2 are separated. The pure CO_2 is either stored in the reservoir or sent to specific industries for utilization

The absorption column in the PCC process captures CO_2 from flue gas using solvent such as Monoethanolamine (MEA), then the gas/solvent mixture (MEA + captured CO_2) is sent to the stripper for solvent regeneration. Thus, solvent regeneration requires energy that is provided by the steam resulted for electricity generation (Rubin et al., 2012; Wang et al., 2011). The energy demand for solvent regeneration is one of the process constrains (House et al., 2009). However, if the heat of the flue gases from combustion chamber is recovered to be used in the stripper section, it will improve the power plant efficiency (Hanak et al., 2014).

The PCC process challenges are, the low CO_2 concentration in the flue gas, the low CO_2 partial pressure that reduces the capture efficiency and increases the energy demand (Wang & Song, 2020; Younas et al., 2016). Furthermore, the removal of the flue gas pollutants such as SO_2 and NO_x increase the energy demand and reduce the power plant efficiency (De Visser et al., 2008).

The estimated cost of CC by the three technologies is shown in **Table 1**, capture by oxy-fuel combustion technique has the highest operating cost due to the high purity of oxygen needed for the process and other process specifications. Although the pre-combustion requires the lowest cost to capture, the invisibility of pre-combustion to retrofit into an existing power plant increases the potential of post-combustion capture to be the optimum technique for CC from power plant.

Table 1 Cost estimated for the CC technologies and viability to be retrofitted to a power plant

Technology	Capturing efficiency (%)	Capture Cost (USD/t)	Visibility to be retrofitted into power plant	Reference
Pre-combustion carbon capture	90	34 – 63	• Difficult fitting without process modification if the power plant is working • Visible only for new power plant	(Kheiririk et al., 2021; Rubin et al., 2015)
Oxy-fuel combustion carbon capture	90	52 - 257	More studies to find the possibility of retrofitting	(Dinca et al., 2018; Kheiririk et al., 2021; Rubin et al., 2015)
Post-combustion carbon capture	90	46 – 74	• It can be easily fitted into power plant if the required place is available • Visible for both new and working/old power plant	(Jiang et al., 2021; Kheiririk et al., 2021; Vega et al., 2019)

1.2.3.4 Post-combustion carbon capture options

The PCC capture of CO₂ is the most effective method for reducing CO₂ emission from power plants (Sun et al., 2020). The PCC process captures CO₂ emitted from burning of fossil fuel and it is considered as the best option to be retrofitted to the existing power plants (Paraschiv & Paraschiv, 2020). The flue gas leaves the stuck near the atmospheric pressure and at temperature ranging between 50-150°C, while the CO₂ concentration in the flue gas is in the range 10-15% by volume (Chen et al., 2013). The three most efficient PCC techniques for separating CO₂ from the flue gas are

using adsorption, chemical absorption, and membrane techniques (Zhang et al., 2018) as shown in Figure 5.

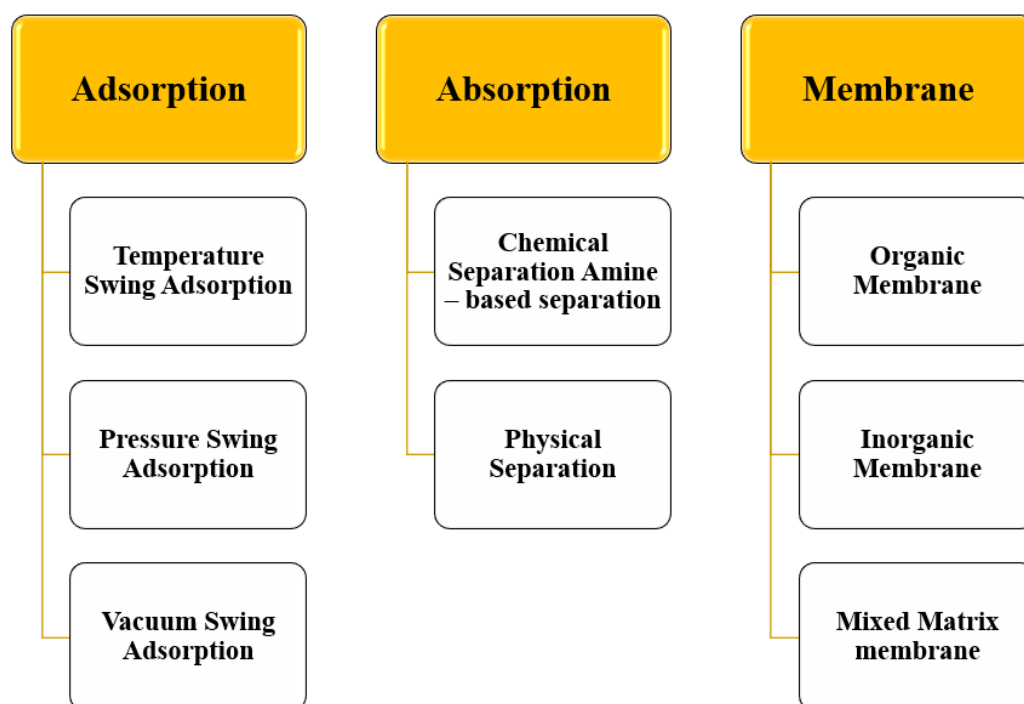


Figure 5 Various options of PCC processes.

1.2.3.4.1 Adsorption as a separation technique

The adsorption method is used to separate CO₂ from flue gas by passing it through a solid surface in a packed column filled with solid at moderate temperature in the range of 26–65°C (Leung et al., 2014; Mukherjee et al., 2019). The adsorption method is classified either as temperature swing, pressure swing, or vacuum swing adsorption that separates CO₂ from flue gas using solid/sorbent located inside a vessel either as fixed bed or fluidized bed (Chao et al., 2021). Comparison between the three techniques according to their ability to deal with the impurities in the flue gas, the required energy demand, and other conditions to improve the process technical and economic challenges are described in Table 2.

The adsorption by temperature swing method works at low temperature following van der Waals force's principles, once the sorbent reaches the saturation condition, regeneration of the sorbent is required at high temperature to enable CO₂ separation from the sorbent, followed by resending the generated sorbent for further CO₂ capture. The advantages of the adsorption process is the low process temperature, low operating cost, and easy integration to any existing power plant (Kuramochi et al., 2012). On the other hand, the adsorption by pressure swing works by adsorbing

CO₂ at high pressure and releasing it at low pressure during the regeneration stage (Ishibashi et al., 1996). The vacuum swing adsorption is the most applicable option at industrial level, the adsorption is carried out under normal pressure and the regeneration is carried out under vacuum where one column is used for adsorption and desorption (Kikkinides et al., 1993). To achieve the capture efficiency, dried and treated flue gas should be maintained for both pressure swing and vacuum swing adsorption that increases the cost of capture process (Zhang et al., 2008).

Table 2 Description of the classification of adsorption separation techniques

No.	Adsorption Separation Technique	Impurities in Flue Gas	Energy Demand	Requirement
1	Temperature Swing Adsorption (TSA)	- Removal of impurities from flue gas is not strict - Impurities could be removed by the adsorbent and later separated from adsorbent at higher temperature	High energy demand around 140°C is the regeneration temperature	Development of sorbent that has low regeneration temperature and high capacity for CO ₂ capture
2	Pressure Swing Adsorption (PSA)	Impurities must be removed from flue gas before adsorption	- Low temperature for adsorption step - High energy demand for pressurization	Hot flue gas is required to reduce the process energy demand
3	Vacuum Swing Adsorption (VSA)	Impurities must be removed from flue gas before adsorption	- Low temperature for adsorption step - High energy demand to achieve vacuum stage	-

Data summarized from literature reports (Jiang et al., 2020; Su & Lu, 2012; Wang et al., 2013)

The PCC by temperature swing adsorption takes place between 40–60°C while the regeneration of the sorbent at two temperatures, 120°C for separating CO₂ from sorbent and 160°C for removing any other impurities (Tlili et al., 2009). Furthermore, the process constraints are the large amount of air needed for heating the sorbent for regeneration stage, the low heat capacity of air, and the high amount of electricity required if the bed is heated electrically. The operating temperature, the energy consumption, and the amount of CO₂ captured, are important parameters to be considered during the design of adsorption process (Chao et al., 2021). For example, the higher the amount of CO₂ to be captured, the higher energy for regeneration is required (Clausse et al., 2011).

1.2.3.4.2 Absorption separation technique

Absorption separation involves physical and chemical techniques, however, the PCC process using chemical absorption is considered the most feasible and easily installed to power plant (Feron, 2010; Ferrara et al., 2017; Figueroa et al., 2008). Although, the technique of chemical absorption is similar to the adsorption separation, the principles are different where liquid is used as a separation media for chemical absorption compared to solid is used for adsorption separation (Spigarelli & Kawatra, 2013). Furthermore, the solubility of the CO₂ in the solvent follows Henry's law that states "*the solubility of the gas is directly proportional to the partial pressure of the gas*" (Maniarasu et al., 2021).

The chemical absorption involves a counter current flow of the flue gas containing CO₂ and the lean solvent in the absorber column at a temperature range of 40–60°C while the regeneration of the rich solvent that lift the absorber column carrying the captured CO₂ takes place between 120 – 160°C within the stripper column (IPCC, 2005; Olajire, 2010; Pires et al., 2011), **Figure 6** shows the process description of the PCC using chemical absorption.

The amine-based solvents for absorption processes are extensively in use due to their characteristic features like, low capital cost and high CC efficiency (Yang et al., 2008). Although the solvent regeneration energy, solvent loss and degradation are hurdles and highly challenge the process implementation, hence, research and development are in a continuous progress to resolve these challenges (Balraj et al., 2022). Several absorption techniques are commercially introduced (Kishimoto et al., 2009; St  phenne, 2014) aiming to reduce the energy demand and enhance the loading capacity using optimization technology. The advantages and

disadvantages of PCC commercial absorption capture techniques are tabulated in **Table 3**. Thus, Process Intensification (PI) approach is implemented in the PCC absorption option, the approach directs to make the process more efficient, reduce the energy consumption, reduce the cost, improve safety and environmental performance (Haase et al., 2022).

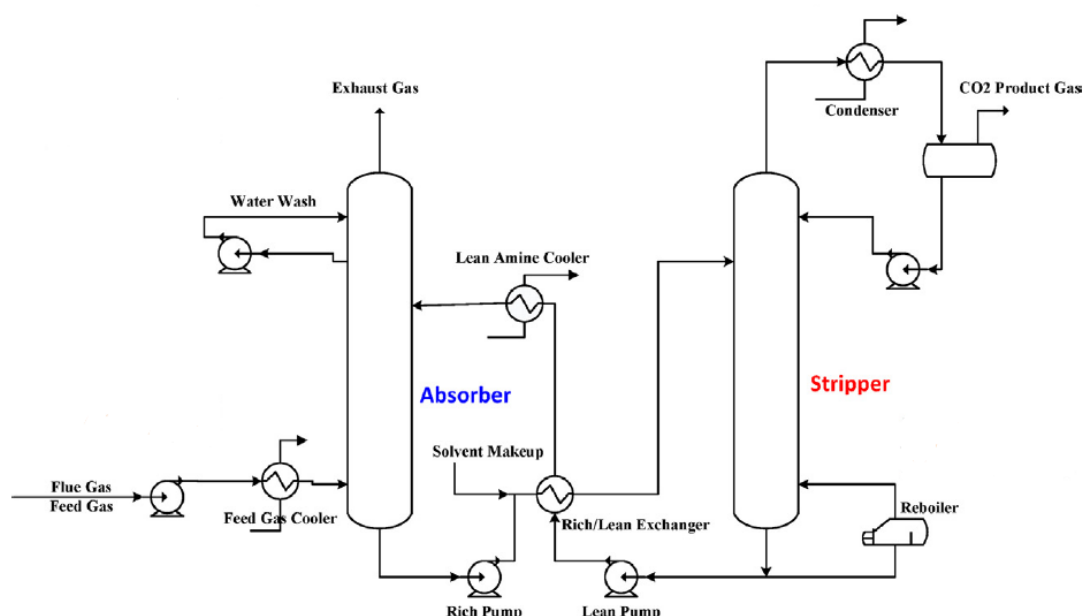


Figure 6 Post-combustion CC diagram using chemical absorption. (Alalaiwat & Khan, 2023)

Table 3 Advantages and disadvantages of highly commercial chemical absorption techniques

No.	Chemical Absorption Technique ^a	Advantages	Disadvantages
1	The KMALC process ^b	• Use low amount of MEA 15-20% • Low regeneration energy	• Use two absorption columns in parallel, high cost
2	The fluor EFG + process ^c	• Designed to reduce the regeneration energy and capital cost. • Can be used under low pressure	• High concentration of MEA is required. • Required of adding inhibitor to reduce MEA corrosive nature
3	KM-CDR technology ^d	• Low CO₂ capture costs for oil industries • Energy efficiency is improved	• The utilized solvent is corrosive. • Low CO₂ loading capacity

4	Aqueous ammonia (NH ₃) process	• NH₃ is solvent that is cheap and has high CO₂ absorption capacity. • Less corrosive and low possibility to degradation	• Possibility of formation the hazardous carbamate • High NH₃ volatility • Low operating temperature that affects CO₂ capture efficiency
5	Chilled ammonia process	• Absorb CO₂ at low temperature that helps to reduce the losses of NH₃ • Low energy for solvent regeneration	• Needs multiple absorber vessels. • Low absorption rate
6	Dual alkali absorption	• Sodium Chloride (NaCl) is added to enhance capture efficiency	• Pre-treatment of the flue gas is required to remove impurities
7	Strong alkali absorption	• Alkali metal-based oxides are used as solvent. • High regeneration temperature that reduces the possibility for solvent degradation	• Low absorption rate of CO₂ • Extensive equipment size is required

Data summarized from literature reports (*Barchas & Davis, 1992; Chao et al., 2021; Scherffius et al., 2013*)

^aThe commercial absorption separation techniques, designed to capture CO₂ from flue gas either to be stored or to be utilized.

^bTechnology developed by Kerr-McGee Chemical Corporation, CO₂ recover technology to be used for food production

^cThe PCC was designed by Fluor using the Econamine FG Plus technology to capture 90% of CO₂ from power plant.

^d The technology developed by collaboration between Mitsubishi Heavy Industries and Kansai Electric Power to capture CO₂ in oil industries.

1.2.3.4.3 Membrane separation technique

Membrane separation of CO₂ capture is relatively new technology using semi-permeable membrane to separate two phases, based on the effect of diffusion, adsorption, molecular sieve action, or ionic transport (Mondal et al., 2012; Yuan et al., 2016), shown in **Figure 7**. In fact, the selection of membrane is an important step to maintain the required capture efficiency, hence, the life-time, operational and material cost, capture performance are important factors of membrane specification that needs to be studied before final selection of the membrane (Spigarelli & Kawatra, 2013). The

specifications of the most applicable membrane for capture are listed in **Table 4**. The driving force for membrane separation is the pressure and the CO_2 concentration, thus the membrane separation is preferable to be used for pre-combustion process, due to the high concentration and high partial pressure of CO_2 , rather than PCC being operative at low concentration and low partial pressure of CO_2 (Wang & Song, 2020; Zhao et al., 2010).

Below, **Figure 7**, indicates a selected membrane that separates CO_2 from flue gases, the membrane is either porous or semi-porous that allows CO_2 to be separated from flue gases under high pressure and a specific concentration in the mixture.

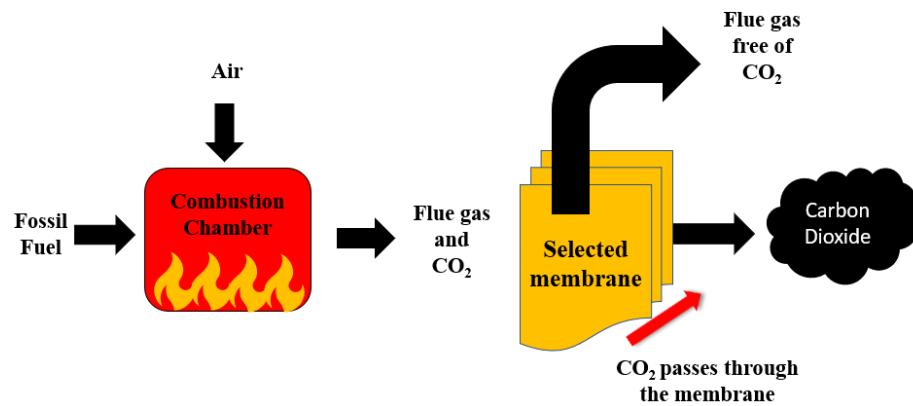


Figure 7 Carbon capture processes using membrane separation technique.

Table 4 Challenges to the most applicable membrane for CO_2 capture.

No.	Type of Membrane	Materials Utilized	Challenges of Membrane
1	Polymer membranes	- Cellulose acetate - Polysulfone - Polyethersulfone - Polyimide	- Low solubility of CO_2 in the membrane material - Needs modification of the membrane packing structure
2	Inorganic membranes	- Porous inorganic - Non-porous inorganic	- Low CO_2 permeability - Expensive
3	Mixed matrix membrane	Composite structure of polymers and inorganic fillers	- Needs to ensure a good adhesion between the membrane layers - Expensive

Data summarized from literature reports (Al-Mamoori et al., 2017; Chen et al., 2020; Norahim et al., 2018; Olajire, 2010; Yuan et al., 2016)

Although CO₂ capture by membrane is simple with no involvement of chemicals or regeneration step, the technology is still under development due to certain challenges such as, the high initial pressure, increase mass transfer resistance once membrane is wet, and the high requirement of CO₂ concentration in the flue gas (Wang et al., 2017; Zhao et al., 2016). In addition, a multi-stage of membrane is required to achieve high separation of CO₂ that is attributed to the low practical transmembrane pressure ratio and membrane selectivity (Zhao et al., 2010).

The estimated operating costs for CC using the three PCC technologies are shown in **Table 5**. Among the three, absorption separation technique has the lowest operating cost with acceptable efficiency and has the potential to be the ideal option compared to the others. In addition, research and development is underway on the absorption technique and is expected to enhance the utilization of this technology once the process drawback is overcome.

Table 5 Comparison of cost estimated for the PCC technologies and the potentials for cost reduction

PCC Technology	Capture efficiency (%)	Capture Cost (USD/t CO ₂)	Potential to reduce the cost	Ref.
Adsorption separation technique	88 – 95	89.66	Needs to Select a source of electricity with low cost	(Cheng et al., 2021)
Absorption separation technique	85 – 90	44 - 64	- Needs to reduce the sizes of absorber and stripper - Needs to select solvent has low cost and high efficiency	(Amrollahi et al., 2011; Li et al., 2016; Yun et al., 2020)
Membrane separation technique	70 – 90	80.46	Needs to enhance the research and development of the technology to be visible for application to power plant	(Zanco et al., 2021)

1.3 Problem Statement

As mentioned earlier, power plants are the key to meet the growing global energy demand, and the process supporting the power generation, using fossil fuels, is subjected to various operational challenges and environmental issues (IEA, 2020). The PCC process appears to be a significant portion of power plant that enables reduction of the plant environmental issues and the promising solution to reduce the emissions of CO₂ (Rochelle, 2016). Despite various implementations of PCC, the energy consumption and the environmental impact of the processes are still considered as process constraints (da Cruz et al., 2021; Zhao et al., 2021). A high energy demand for PCC process results from the large steam required for solvent regeneration specifically for reboiler unit (IEA, 2011). For example, the standard PCC process with 30 wt% MEA consumes energy in the range of 3.2 to 5 GJ/tCO₂ (Canepa et al., 2012; Mimura et al., 1995). Several studies suggest that solvent characteristics, concentration,

and CO₂ lean loading have directly affected the reboiler duty and the regeneration energy (Gervasi et al., 2014; Otto, 2011). Some researchers suggested process configurations to reduce the energy demand such as stripper split feed, lean vapor compressor, and changes in other operating parameters (Gervasi et al., 2014). However, the process configurations to reduce energy demand, increases the process capital cost, for instance ,reducing the capture cost by 10% from 58.1\$/t-CO₂ due to process configurations, increases the capital cost from \$312.4 to \$325.2 (Khalilpour & Zafaranloo, 2020).

The piperazine (PZ) was considered as an alternative solvent because of its superior properties to MEA, by the ability to capture CO₂ at higher capacity with lower energy for regeneration and lower volatility rate (Freeman et al., 2010). However, the identified high degradation rate of PZ during the PCC process and the precipitation of aqueous PZ solution at low temperature less than 20°C that caused an impact on environment and human health, is a matter of concern (Rabensteiner et al., 2015). The weaknesses of PZ have encouraged investigation of other solvent options to enhance the PCC efficiency and reduce energy demand. This opened the room for further investigating to the effectiveness of solvent blends (Dash et al., 2014; Nwaoha et al., 2017). For example, an amine has a high absorption capacity such as methyldiethanolamine (MDEA) blends with a solvent having high rate of reaction such as PZ enhances the capture efficiency (Chakravarty et al., 1985). The low regeneration energy and high-capacity loading of MDEA encouraged the investigation for activator, where PZ appears to be the optimum to accelerate the rate of reaction of MDEA with CO₂ (Alalaiwat & Khan, 2023). However, the benefits of blended amines solvents depend on the individual concentration of each solvent (Sakwattanapong et al., 2005). Consequently, the development of an optimization model to investigate the synergistic effect of MDEA/PZ to reduce regeneration energy and enhance capture efficiency by identifying the optimum amount of the individual solvents is highly recommended.

The PCC process shows reliability to reduce the emissions of CO₂ from the stationary source, however, there is a worry about process effect on ecosystem and human health that needs to be further studied to evaluate its environmental impact (Branco et al., 2013). The degradation of utilized chemicals during PCC process is the most serious issue because of the ability of these chemicals to cause corrosion of the equipment or influent from process with chemical waste or wastewater (Matin & Flanagan,

2022), that needs further attention to include treatment units (Pehnt & Henkel, 2009).

Indeed, to enhance the utilization of specific process it needs to be investigated from technical and environmental point of view. Several environmental assessment were introduced such as environmental impact assessment, and life cycle assessment. Life Cycle Assessment (LCA) is evaluate the environmental impact of process throughout the life cycle from first stage to disposal, furthermore, its considered the most implementing methodology to evaluate the environmental performance of PCC once retrofitted to power plant due to the methodology ability to examine the impact in each stage and provide the carbon footprint (Boot-Handford et al., 2014; Giordano et al., 2018; Koornneef et al., 2008; Pehnt & Henkel, 2009). Hence, it's highly recommended to find out the environmental impact of PCC process using the best technical performance option that helps decision-maker to widely select the strategy for utilizing carbon capture.

1.4 Objectives of this study

This PhD research viewed the importance of the PCC and severity of CO₂ emission that aims to:

- Develop HYSYS model for PCC process to capture the emitted CO₂ from Power Plants.
- Develop LCA scenario of the PCC model to assess the environmental impact of the process using SimaPro application/software.
- Compare the LCA with different alternative solvents used for the developed PCC.

With these aims this study would strive to achieve objectives such as:

- To identify the optimum solvent and its concentration to be used for capturing CO₂
- To develop and validate a simulation model for PCC process.
- To model a LCA for the model PCC process with regard to the two selected solvents.
- To compare the LCA of PCC of the two selected solvents relative to the power plant without PCC.

1.5 Research questions

This work strives to answer the following questions:

- *Which process variables have a strong influence on the energy demand for solvent regeneration*
- *Is the PCC process the optimum CC technique to be retrofitted to power plant?*
- *What are the effects of solvents synergism to the PCC energy demand and process efficiency?*
- *What is the environmental life cycle impact of PCC process using solo solvent and blended solvents?*

1.6 Sustainable development goals related to this study

In 2015 Paris agreement is set in which the United Nations (UN) developed 17 goals to support countries to maintain sustainability that are designed based on three dimensions including, environment, economy, and society known as SDG (SDG, 2018). All nations are requested to set their plan to achieve their SDG target taking in to consideration the UN theme of “no one will be left behind” (Scheyvens et al., 2016; SDG, 2018). To achieve the SDG targets, indicators are set and link to each SDG, furthermore, to monitor and follow up the progress of countries toward SDG, UN set a follow-up and review mechanism that will help countries to assess their progress toward the target of each goal (Stevens & Kanie, 2016).

Climate action the SDG number 13, that considers the SDGs needs further actions and more progress to reduce the impact of climate change across different sectors. Climate change has altered the patterns of weather, raised atmospheric temperature, raised ice melting which are considered as a threatening for life of human beings and other organisms (Church & White, 2006; IPCC, 2013). Without action, the atmospheric temperature is expected to reach 3°C by the end of this century (Doni et al., 2020). According to UN, the climate change impacts are not limited to a specific area or a country, collaboration must be established to implement solutions specifically by shifting to low carbon economy (IPCC, 2022). The SDG 13 contains 5 targets need to be considered by countries across the globe as listed in **Table 6** (SDG, 2022).

Table 6 Sustainable Development Goal 13–5 main targets

Target No.	Description
13.1	Strengthen resilience and adaptive capacity to climate – related hazards and natural disasters in all countries
13.2	Integrated climate change measures into national policies, strategies and planning
13.3	Improve education, raising awareness and human and institutional capacity on climate change mitigation, adaptation, impact reduction and early warning
13.4	Implement the commitment undertaken by developed – country parties to the United Nations Framework Convention on Climate Change to a goal of mobilizing jointly \$100 billion annually by 2020 from all sources to address the needs of developing countries in the context of meaningful mitigation actions and transparency on implementation and fully operationalize the Green Climate Fund through its capitalization as soon as possible
13.5	Promote mechanisms for raising capacity for effective climate change – related planning and management in least developed countries and small island developing states, including focusing on women, youth and local marginalized communities

Climate change has a critical impact on several important sectors. For example, a reduction in the crop harvest by 70% by 2030 is expected as an adverse effect (Challinor et al., 2014), and it severely affects the agriculture and the food system compared with other sectors (Porter et al., 2014). Hence, the integration effect between the SDGs can help reducing the climate change consequences, for example, increasing the contribution of renewable energy could help to reduce the utilization of fossil fuels that are considered the highest emitter of greenhouse gases. On the other hand, improvement in health will increase due to the reduction in emissions and more job opportunities will be created due to the investment in the renewable energy (Doni et al., 2020). The UN to reach carbon free future (UNEP, 2020b) has emphasis on the importance of a circular carbon economy framework that takes into account the way to deal with excess of CO₂ emissions through the following stages:

- Reduce: which could be achieved by improving the contribution of renewable energies and alternative energy sources and enhancing energy efficiency.
- Reuse: which could be achieved by using CCU.
- Recycle: which could be achieved by recycling carbon to be used for energy purposes such as bioenergy and hydrogen.

- Removal: which could be achieved by using Carbon Capture and Storage (CCS) or Direct Air Capture (DAC).

Accordingly, this study is highly linked to the SDG and specifically contributes to resolving the issues related to goal 13 as illustrated in Table 7.

Table 7 SDG goals, targets, and indicators associated with this research

SDG	SGD Goals	SDG Target	Indicator
13 Climate Action	Take urgent action to combat climate change and its impact	13.2 Integrate climate change measures into national policies, strategies, and planning	13.2.1 Number of countries with nationally determined contributions, long-term strategies, national adaptation plans and adaptation communications, as reported to the secretariat of the United Nations Framework Convention on Climate Change
			13.2.2 Total greenhouse gas emissions per year

1.7 Significant of the study

This study focuses on PCC process that is retrofitted to power plant, furthermore, based on the literature review presented in chapter 2, the significant contribution of this thesis are as follows:

- It contributes to reducing the emissions of CO₂ from the power plant, classified as the highly emitter source, where CO₂ as a major contributor of global warming and climate change. Furthermore, the study supports the transition to cleaner energy resources that will foster the movement to reach net zero emissions.
- A multi-regression formula to be used to estimate the optimum energy demand and the amount of absorbed CO₂ per utilized solvent was developed. The formula can be used to predict the CO₂ rich loading and regeneration energy for PCC process with standard configuration using solo solvent. However, the developed formula can be used to estimate the reduction of the regeneration energy once new configurations are utilized.
- Extensive studies related to PCC have been conducted to investigate the energy demand and process efficiency using MEA, very few

studies have addressed the effects of blended solvent (multi-solvents), and almost no study investigated the optimum concentration of multi-solvents by modeling and simulation based on pilot plant data. The MDEA and PZ are considered the best solvents with synergistic effect to capture CO_2 with low regeneration energy compared to pure MEA. The finding of the PCC modeling and simulation using MDEA/PZ could be used as a basis for utilization of PCC in power plant and industries. Furthermore, exploring the PCC model based on MDEA/PZ blended system could be used to estimate the economic part of the process and to be used for further process's environmental assessment.

- A tremendous investigations have applied to the LCA to study the environmental impact of specific scenarios. The PCC retrofitted to power plant using MEA was the only identified in the literature, moreover, most studies deal with data from literature and limited to real or pilot plant process. The LCA of PCC using MDEA/PZ based on pilot plant data is the first to be investigated and it shows a reduction of environmental impact compared to PCC with MEA. The findings of the LCA show the possible areas of environmental impact that need further consideration by issuing policy or framework during the utilization of PCC. It is important to note that the PCC with MDEA/PZ stands as a favorable option, while the research and technological advancements will continue to shape the CC process.

1.8 Limitation of the study

The focus of this piece of work is the modeling, simulation, and analysis of the optimum solvent for PCC process from technical and environmental view, hence the study is limited to the following:

- To CO_2 capture using MEA and blended solvent containing MDEA/PZ.
- Modeling of power plant facilities, flue gas desulphurization, the captured CO_2 transportation and storage are not covered in this study.
- The multi-regression looks at the PCC significant variables for solo solvent while the blended solvents are not covered in the multi-regression analysis.
- The PCC model development, model validation, and technical analysis for solvent concentration for a constant flow rate using both

MEA and MDEA/PZ through HYSYS. The process configuration is not covered.

- The environmental assessment using the LCA methodology for the optimum PCC model developed is excluded the construction, infrastructure, transportation, and CO₂ storage from this study.
- All studies performed in this thesis are based on study-state models.
- The economic assessment of PCC process is not covered in this study.

1.9 Structure of the thesis

The structure of four chapters in this thesis is as: **Chapter 2** focuses on the review of the existing work related to the solvent-based PCC using MEA and synergistic solvent system for standard PCC configuration and developed PCC configuration. Summary of the review is included at the end of the chapter.

Research methodology using Multi-regression analysis for standard PCC process retrofitted to power plants using SPSS, HYSYS model, and LCA methodology are presented in **Chapter 3**. It includes the key process variables of the PCC process.

In **Chapter 4**, the results from the multi-regression analysis, HYSYS model, and LCA are presented followed by the conclusion and recommendations for future work.

1.10 Definition of terms

The scientific terms used in this study are defined as follows:

- **Power plant:** is a facility designed for generating electric power specifically by converting specific fuel, renewable or non-renewable, to electricity. The facility require huge amounts of water that change from liquid to steam to be used for running a turbine. (IEA, 2011)
- **Flue gas:** is the gas stream resulting from fossil fuels combustion in a power plant, that contains various pollutants such as CO₂ (IPCC, 2018).
- **Greenhouse gases:** the atmospheric gases responsible for rising of atmospheric temperature and climate change because of their ability to absorb infrared radiation and retain heat contents. The major greenhouse gases are carbon dioxide, methane, and nitrous oxide. They are highly emitted from human activities (UNEP, 2021).

- **Global warming potential:** an index that presents the combined effect due to the presence of greenhouse gases in the atmosphere and their effectiveness to absorb the outgoing infrared radiation (UNEP, 2021).
- **Carbon dioxide equivalent:** is a measure used to present the total global potential of greenhouse gases in terms of the amount of carbon dioxide. It provides a standardized way of comparing the effect of different greenhouse gases (UNEP, 2021).
- **Carbon capture and storage:** refers to the technology that is designed to mitigate the emissions of CO₂ from industries and energy sources that involves three stages namely, capture CO₂ from the source, followed by transportation to the storage area, and ends up with storing the captured gas either in the depleted oil or gas reservoirs or deep in ocean (IPCC, 2005).
- **Net zero emissions:** is a state where the total amount of GHGs emitted into the atmosphere is balanced or equal to zero either by removal or offset of these gases that helps to reduce the impact of climate change and global warming (IEA, 2022).
- **Post-combustion capture:** it's a technology that refers to the capture of emitted CO₂ from power plants using specific solvent. The solvent can work by either physical or chemical separation technique where the energy demand, cost, and solvent degradation are the main process challenges (IEA, 2007).
- **Solvent:** is a chemical compound that can be mixed up with water to form a homogenous solution. Under certain conditions of temperature and pressure, that solvent can be combined with another compound such as CO₂ to form a mixture that is stable under certain conditions and can be separated at certain temperature (IEA, 2007).
- **Absorption:** is the first stage in PCC process where the selected solvent takes up CO₂ from flue gases forming a solution. This stage is crucial for the process efficiency (IEA, 2007).
- **Stripping:** this is the second stage in a PCC process where the solution of solvent and CO₂ is separated. The separated solvent is recycled to the absorption stage while CO₂ is compressed and sent to storage. This stage requires the highest energy of the process (IEA, 2007).
- **Capture efficiency:** it is the amount of CO₂ captured in PCC process compared to the total emitted CO₂ from a power plant. The higher

capture efficiency indicates that the PCC process is running in the optimum level (IPCC, 2005).

- **Energy penalty:** the energy that is required for functioning the PCC unit being provided either by the power plant or external source. The amount of energy penalty is highly dependent on the power plant efficiency and considered one of the PCC challenges (IEA, 2015).
- **Life cycle assessment:** an assessment that follows ISO 14040 steps and considers the environmental impact of the entire process and its output that provides a holistic view of the implementation of sustainability. The stages of life cycle assessment are goal and scope, life cycle inventory, life cycle impact assessment, and interpretation (ISO, 2006).

Chapter 2

Literature Review

Literature Review

2.1 Overview

This chapter aims at reviewing the progress of research activities of experimental, modelling, simulation, and LCA of CC by solvent-based PCC using both standard and advanced processes. A review of the current pilot plant and the configuration options are provided in section 2.2. The principles of the PCC modelling, equations, process variables, and the performance of PZ, MEA, and synergetic of MDEA/PZ is discussed in section 2.3. The LCA and environmental impact of PCC studies are reviewed in section 2.4. The summary of the literature review is presented in section 2.5.

2.2 Solvent-based PCC process technology worldwide

2.2.1 Review of existing PCC pilot plants

Research and development pertaining to CC expanded rapidly which enabled several pilot plants for PCC around the world to be established. The brief description of the best research of different pilot plants is investigated in reasonable detail (Wang et al., 2011). The work of some authors (Rubin et al., 2012) described the PCC pilot projects worldwide, they are either operating or in design and construction stage. Indeed, the purpose of starting with pilot plant is to ensure that all the issues related to the process are resolved before the full-scale commercial plant design and operation, that issues are identified in the research of Gelowitz and co-workers (Gelowitz et al., 2013). In later stage, technical and economic analysis of solvent-based PCC retrofitted to a real power plant is investigated (Lungkadee et al., 2021) wherein the optimum process conditions and the required cost is identified. Recently, a comprehensive review is carried out (Nessi et al., 2021) containing around 37 solvent-based PCC pilot plant locations worldwide and the performance of different solvents, process configuration, process variables that directly affect the process performance have been analyzed. Several PCC pilot plants and the related key information are shown in **Table 8** for easy comparison and reference.

Table 8 Summary of the identified based-solvent pilot plants

Pilot plant	Location	Flue gas flow rate	Solvent	CO ₂ capacity (t/day)	PCC design (Standard/Advanced/Developed)	Ref.
Separation Research Program ^a (SRP)	University of Texas at Austin (USA)	600–1500 m ³ /h	30 wt% MEA, 40 wt% PZ and K ₂ CO ₃ promoted with PZ	3	Standard and Flue gas prepared by the research group	(Chen et al., 2013; Dugas, 2006)
National Carbon Capture Centre (NCCC)	Alabama (USA)	1656–2376 kg/h	30 wt% PZ	10	Standard and the flue gas is provided from power plant that diluted to reach the required CO ₂ concentration	(Rochelle et al., 2022)
CSIRO Taronga ^a	Queensland (Australia)	500–900 kg/h	30 wt% MEA, 40 wt% PZ	2.7	Standard and the flue gas is provided from power plant	(Akula et al., 2021; Cousins et al., 2011b; Cousins et al., 2011a; Li et al., 2016)
International Test Center (ITC)	University of Regina (Canada)	200 Nm ³ /h	30 wt% MEA, MEA/MDEA	1	Standard and Flue gas prepared by the research group	(Edali et al., 2009)
Laboratory of Engineering Thermodynamic	University of Kaiserslautern (Germany)	30–110 kg/h	30 wt% MEA, new solvents	-	Standard and Flue gas prepared by the research group	(Mangalapally & Hasse, 2011; Notz et al., 2012)
CO ₂ Separation Plant	Durnroht (Austria)	50–120 m ³ /h	30 wt% MEA, 37.6 wt% PZ, promoted AMP/PZ	0.6	Standard and the flue gas is provided from power plant	(Rabensteiner et al., 2015)
The Esbjerg CESAR EU project	Esbjerg power station (Denmark)	5000 Nm ³ /h	30 wt% MEA,	24	Standard and the flue gas is provided from power plant	(Faber et al., 2011; Gaspar et al., 2017)
Center for Applied Energy Research	University of Kentucky (USA)	-	30 wt% MEA, advanced solvent (A and B)	-	Developed with two stripper columns and the flue gas is provided from power plant	(Frimpong et al., 2019)

^a The experimental data from the pilot plant was used for model validation in this thesis.

The experimental data of the reviewed pilot plants is used as a source for model validation. The developed model based on the pilot plants has been used for investigating and testing various PCC process improvements such as evaluation of the performance of solvent to capture CO₂ (Bishnoi & Rochelle, 2000; Freeman et al., 2010; Hwang et al., 2019; Liu et al., 2022), modifications of process configuration and energy demand (Jiang et al., 2021; Mofidipour & Babaelahi, 2020; Oh et al., 2020; Oh et al., 2018; Walters et al., 2016), finally the environmental impact of the process (Dai et al., 2012; Liang et al., 2015; Supap et al., 2011). Indeed, the successful

achievement of the pilot plant test and the demonstration results will enable the high commercial implementation of solvent-based PCC process.

2.2.2 Commercial operation of solvent-based PCC process

The PCC has gained greater attention and has been demonstrated at different level to capture CO₂, for example, Petra Nova (PN) project in USA that captures 1.6 Mt/year CO₂ using Mitsubishi Heavy Industries (MHI) KM CDR process, and Boundary Dam (BD) at SaskPower in Canada that captures 1 Mt/year, are the biggest PCC projects where the CO₂ captured from the two projects is used to enhance oil recovery (Liang et al., 2015; Montañés et al., 2017). The advantages and disadvantages of the commercial techniques are shown in Table 9.

Table 9 Advantages and disadvantages of highly commercial chemical absorption techniques.

No.	Chemical Absorption Technique ^a	Advantages	Disadvantages
1	The KMALC process ^b	• Use low amount of MEA 15-20% • Low regeneration energy	• Use two absorption columns in parallel, high cost
2	The fluor EFG + process ^c	• Designed to reduce the regeneration energy and capital cost. • Can be used under low pressure	• High concentration of MEA is required. • Required of adding inhibitor to reduce MEA corrosive nature
3	KM-CDR technology ^d	• Low CO₂ capture costs for oil industries • Energy efficiency is improved	• The utilized solvent is corrosive. • Low CO₂ loading capacity
4	Aqueous ammonia (NH ₃) process	• NH₃ is solvent that is cheap and has high CO₂ absorption capacity. • Less corrosive and low possibility to degradation	• Possibility of formation the hazardous carbamate • High NH₃ volatility • Low operating temperature that affects CO₂ capture efficiency
5	Chilled ammonia process	• Absorb CO₂ at low temperature that helps to reduce the losses of NH₃	• Needs multiple absorber vessels. • Low absorption rate

		• Low energy for solvent regeneration	
6	Dual alkali absorption	• Sodium Chloride (NaCl) is added to enhance capture efficiency	• Pre-treatment of the flue gas is required to remove impurities
7	Strong alkali absorption	• Alkali metal-based oxides are used as solvent. • High regeneration temperature that reduces the possibility for solvent degradation	• Low absorption rate of CO₂ • Extensive equipment size is required

Data summarized from literature reports (Barchas & Davis, 1992; Chao et al., 2021; Scherffius et al., 2013)

^aThe commercial absorption separation techniques, designed to capture CO₂ from flue gas either to be stored or to be utilized.

^bTechnology developed by Kerr-McGee Chemical Corporation, CO₂ recover technology to be used for food production

^cThe PCC was designed by Fluor using the Econamine FG Plus technology to capture 90% of CO₂ from power plant.

^d The technology developed by collaboration between Mitsubishi Heavy Industries and Kansai Electric Power to capture CO₂ in oil industries.

2.3 Solvent-based PCC for CO₂ capture

2.3.1 Principles of modelling PCC process

The PCC using chemical absorption technique involves two operations which take place simultaneously, they are chemical reaction and gas-liquid mass transfer, taking place in both absorber and stripper columns. The equilibrium and non-equilibrium approaches are considered in the PCC process; hence, rate-based model is considered for the process modeling rather than equilibrium stage model (Okon et al., 2015). Furthermore, the rate-based model describes the heat and mass transfer between the liquid and gas within the PCC process rather than assuming equilibrium between the liquid and gas phases (Lawal et al., 2009). In addition, the reversible reaction of the solvent with CO₂ is taken place rapidly and assuming reaction under equilibrium will fail the PCC process (Lawal et al., 2012). The rate-based approach is used to model the rate of the chemical reactions and the rate of mass and heat transfer of multi-component within the columns (Okon et al., 2015). The mass transfer model uses two-film theory following Maxwell-Stefan formulation that uses to find the mass fluxes of

the liquid and gas phases components as shown in **Figure 8** (Lawal et al., 2009).

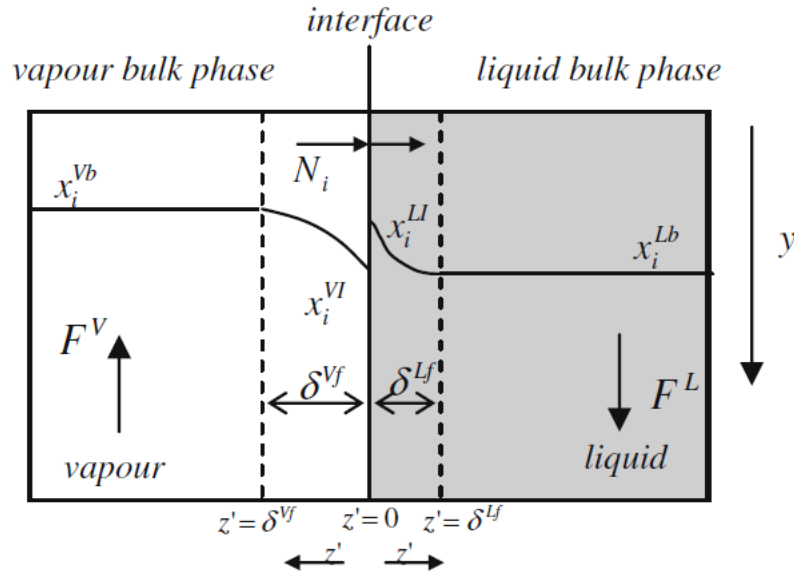


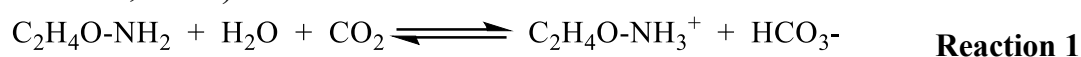
Figure 8 Liquid and vapor/gas films and interface (Lawal et al., 2009).

The chemical reaction of CO₂ with solvent specifically amine is described via three mechanisms those are Zwitterion mechanism, single-step mechanism, and carbamic acid reaction mechanism. These mechanisms are studied in detail (Bishnoi & Rochelle, 2000; Derks et al., 2006; Littell et al., 1992). The rate of reaction of PZ and MEA is fast compared to MDEA, this can approximately assume the reaction reaches equilibrium for PZ and MEA while reaction kinetics of MDEA needs further exploration (Zhang et al., 2009). For the reaction equilibrium assumption, the reaction kinetic and the rate of reaction are identified using the enhancement factor based on pseudo-first order reaction (Derks et al., 2006).

A rigorous model of PCC process using rate-based model can be established using commercial simulator such as Aspen Plus (Kvamsdal et al., 2009; Lungkadee et al., 2021; Zhang et al., 2009), Aspen Hysys (Jiang et al., 2021; Mostafavi et al., 2021; Øi, 2007), gCCS (X. Wu et al., 2018; Wu et al., 2021), gProms (Lawal et al., 2012; Li et al., 2018), and Matlab (Gaspar et al., 2017) to simulate a better fitting model using the mass and heat transfer, and the chemical reactions. Thus, Aspen Plus and Aspen Hysys were the most utilized commercial simulators according to the published reports.

2.3.2 Simulation studies of PCC process using MEA as solvent

According to Liang et al. (Liang et al., 2015) selection of the optimum solvent considers three aspects namely, CO₂ solubility in the solvent, the reaction kinetics of the solvent with CO₂, and the physical and chemical properties of the selected solvent. Alkanolamine compounds are a highly preferable option, that is derived from NH₃ by replacing one or more of its hydrogen atoms during reaction with ethanol. Hence, alkanolamines are classified as primary, secondary, or tertiary amines according to the number of substituents (Chao et al., 2021). Monoethanolamine is one of the most utilized solvents among the other options, **Reaction 1** shows the chemical reaction that takes place where the low temperature drives the reaction to absorb CO₂ by MEA and forms MEA carbamic acid and the high temperature drives the reaction to separate CO₂ from MEA (Herzog & Golomb, 2004).



Typically MEA is the most used solvent in the PCC process to capture CO₂, and the 30 wt% of MEA is considered the optimum amount and the benchmark solvent for any study (Wu et al., 2020). The studies involved MEA are mainly related to modelling and simulation for open loop system in which absorber and stripper are considered as a single component (Dinca & Badea, 2013; Gaspar et al., 2016; Kvamsdal et al., 2009), the whole system studies either to reduce the energy demand or increase process efficiency (Akula et al., 2021; Frimpong et al., 2019; Le Moullec et al., 2014; Li et al., 2018b), and development of the power plant that PCC process is retrofitted to (Bravo et al., 2021; Davison, 2007; Fu et al., 2021).

Addition of inhibitor to the MEA solution and increase its concentration up to 35 wt% shows that the degradation and corrosive nature of the solvent are reduced (Parker & Folger, 2010; Spigarelli & Kawatra, 2013). The effect of the process variables such as MEA concentration, CO₂ loading, and pressure on stripper column were investigated with reasonable justification (Abu-Zahra et al., 2007b; Abu-Zahra et al., 2007a). The investigation found that at CO₂ loading from 0.25 to 0.33 mol of CO₂ per mol of MEA requires less regeneration energy at high solvent concentrations to achieve 90% CO₂ capture efficiency. For example, the regeneration energy of the process dropped from 3.3 to 3.01 GJ/t CO₂ once the MEA concentration increased from 30 to 40 wt%.

The performance of MEA in PCC process under part-load operation is investigated under different process variables (Akula et al., 2021). The model of the study was based on validated pilot plant experiments and the study shows that the part-load operation increases the energy demand and energy penalty. The polynomial Nonlinear Auto-Regressive with Exogenous (NARX) model was developed (Akinola et al., 2019) using Orthogonal Least Squares (FROLS) algorithm to predict the dynamic response of amine-based PCC process that enables to identify the system outputs depending on the system inputs.

2.3.3 Simulation of PCC process using PZ as solvent

The downside of MEA related to the high capital cost, degrades at temperature higher than 120°C, the solvent itself is highly corrosive, requires high regeneration energy, and the possibility to have an environmental issues in the presence of some gases such as O₂ and SO₂, all these challenges invite investigation for an alternative solvent (Davison, 2007).

Piperazine is a secondary amine by nature, has high ability to absorb CO₂, has comparative high stability, needs lower energy in the stripping column and is more resistant to thermal degradation up to 150°C. Hence, PZ appears to be another alternative for MEA and consequently gained high attention in the scientific community (Freeman et al., 2010; Rochelle et al., 2011).

In the beginning, PZ was used as a promoter because of its ability to form carbamate rapidly with CO₂ (Freeman et al., 2010), then the ability of aqueous PZ to capture CO₂ without being promoted with appropriate amount was tested by several researchers and the results were found satisfactory (Otitoju et al., 2021; Pérez-Calvo & Mazzotti, 2022; Rabensteiner et al., 2015).

Technical and economic assessment of the PCC process at different flue gas composition in the flowrates was studied using Aspen Plus (Pérez-Calvo & Mazzotti, 2022). The study reveals that an increase in the reboiler duty raised the regeneration energy from 2.4 to 2.9 MJ/kg_{CO2} once the concentration of CO₂ in flue gas dropped from 33 to 4% by volume. However, using 35 – 40% of PZ by weight as a solvent reduced the regeneration energy even if the CO₂ concentration in the flue gas dropped to 4% by volume. Besides the PZ concentration, the CO₂ loading, the split ratio in the absorber, the temperature difference in the lean/rich heat exchanger, and stripper pressure have been identified as process constraints

on the technical and economic performance of the PCC process using PZ (Chen et al., 2017; Dubois & Thomas, 2018; Gao et al., 2019; Gaspar et al., 2016; Rochelle et al., 2019).

The PZ was selected as an alternative solvent that needs less energy for the regenerating stage in the stripper compared to the MEA. Rabensteiner and coworkers (Rabensteiner et al., 2015) found that 37.6 wt% of PZ reduced the regenerating energy by 14% with reference to 30 wt% of MEA once the optimum liquid/gas ratio and absorber height is achieved. The optimum amount of PZ (30 – 40 wt%) is more economically viable option to capture CO₂ compared to 30 wt% MEA being explored by Otitoju et al. (Otitoju et al., 2021).

The pilot plant developed by the University of Texas at Austin for capturing CO₂, used PZ to evaluate the performance of advanced PCC process using advanced flash stripper configuration. The test showed that 30 wt% PZ is more efficient than 30 wt% MEA and the energy requirement was reduced by 25% compared to two stage flash configuration where the heat duty reached by 2.1 -2.9 GJ/t_{CO2} (Chen et al., 2017). The oxidation of PZ was tested at CSIRO pilot plant at the Tarong Power Station in Queensland beside the one at the University of Texas. The test showed that the solvent maintained stability for 425 h of process at 125°C and for 421 h of process at 155°C, while the thermal degradation was not observed even after 1700 h of the operation (Chen et al., 2017; Freeman et al., 2009).

The investigation into PZ aerosol in the absorber revealed that the presence of sulfur trioxide (SO₃) in the flue gas is responsible for aerosol formation (Akinpelumi et al., 2019). It was concluded that when aerosol formation is detected, increasing the lean solvent temperature is necessary as a mitigation measure to promote the growth of aerosol, allowing it to reach a size that enables its collection at the top of the absorber column.

2.3.4 Simulations of PCC process using MDEA/PZ as solvent compared to other solvents

The amine-based solvents are most applicable and is an attractive option to capture CO₂ from power plant using PCC absorption technique (Shaw, 2009). For the past decades, scientists were testing the synergetic efficiency of blending amine solvents to MEA or synthesized amines (Rey et al., 2013). The latter option was commercially not preferable due to the

required high cost and uncertainty for large scale applications (Liang et al., 2016).

Although the single solvent such as MEA, MDEA, and PZ is the most utilized options that effectively remove CO₂ (Apaiyakul et al., 2022; Budzianowski, 2015), single solvent encourages to shift to high efficiency synergies raised from solvent blends in future (Liang et al., 2016). The first synergy involved the blend of MEA and MDEA. The MEA has high regeneration energy, while MDEA has a low reaction rate, hence, combining them enhanced the performance behavior of each solvent independently (Budzianowski, 2015). The MDEA is considered another alternative solvent to MEA because of the low energy consumption in PCC (Ma et al., 2021). However, MDEA shows poor ability to capture CO₂ due to the slow kinetic reaction, in which it required to be promoted by other compounds such as PZ (Bishnoi & Rochelle, 2000). The optimum amount of PZ to promote MDEA to maintain high efficiency and lower energy demand, is investigated to find the ideal composition of the two compounds (Hosseini-Ardali et al., 2020; Mostafavi et al., 2021; Zhao et al., 2017). The regeneration energy for the highly efficient solvents are listed in **Table 10** and lowest regeneration energy of the highly utilized solvent is shown in **Figure 9**.

Table 10 The regeneration energy of different solvents using PCC chemical absorption.

No.	Solvent	Regeneration Energy (MJ/KgCO ₂)	Reference
1	EEMPA ^a	2.3	(Jiang et al., 2021)
2	K ₂ Sol ^b	2.8	(Hwang et al., 2019)
3	MEA	4.3	(Hwang et al., 2019)
4	PZ/K ₂ CO ₃ ^c	3.3	(Oexmann & Kather, 2009)
5	MDEA/PZ ^d	2.74	(Zhao et al., 2017)
6	MDEA/PZ	2.76	(Hosseini-Ardali et al., 2020)
7	MEA	3.57	(Hosseini-Ardali et al., 2020)
8	PZ	2.80	(Otitoju et al., 2021)
9	MEA	5.34	(Otitoju et al., 2021)

^a2-Ethoxyethyl-3-morpholinopropan-1-amine, a water-lean solvent.

^bAqueous alkanolamine, one of the water-lean solvents.

^cThe solvent is formed from potassium carbonate that is used to promote PZ to enhance the capture efficiency.

^dThe solvent system is MDEA, promoted by PZ to enhance the capture efficiency.

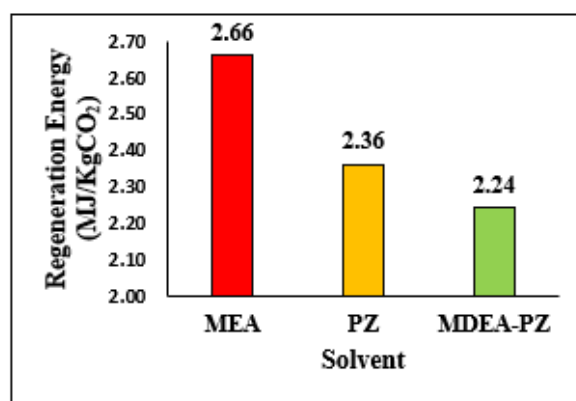


Figure 9 Regeneration Energy for the highly utilized solvents

The study conducted by Hosseini-Ardali and co-workers using Aspen Hysys simulation found that the optimal concentration of MDEA/PZ to enhance capture efficiency and reduce regeneration energy in a standard PCC process lies between 30 – 40 wt% (Hosseini-Ardali et al., 2020). The PCC system was developed based on results obtained from a pilot plant, which were validated and scaled up to be optimum for 550 MW coal fired power plant. The study shows that the regeneration energy for 94% capture efficiency using MDEA/PZ is less than the regeneration energy of 90% capture efficiency using MEA. However, using process configurations such as rich solvent recycle, solvent split flow, and lean/rich vapor compression further reduced the energy demand (Dubois & Thomas, 2018). The summary of PCC simulation using MDEA/PZ is listed in **Table 11** that shows 20 wt% PZ is the highest concentration of MDEA/PZ that needs the lower regeneration duty and lower circulation rates. However, the study of MDEA/PZ performance to capture CO₂ using PCC advanced flash stripper is not yet documented in literature.

Table 11 Summary of PCC simulation using MDEA/PZ solvent system.

No.	Concentration (wt%)	Lean loading (mol CO ₂ / mol solvent)	Rich loading (mol CO ₂ / mol solvent)	Regeneration Energy (MJ/kgCO ₂)	Process Modification	Reference
1	MDEA – 30 PZ – 20	0.1	0.5631	2.24	Intercooling Rich-split Inter-heating	(Zhao et al., 2017)
2	MDEA – 30 PZ – 20	0.19	0.45	2.3	Intercooling	(Tobiesen et al., 2007)
3	MDEA – 10 PZ – 30	0.24	0.75	2.39	Rich vapor compression	(Dubois & Thomas, 2018)

4	MDEA – 40 PZ – 10	0.14	0.37	2.42	Intercooling	(Tobiesen et al., 2007)
5	MDEA – 35 PZ – 15	0.16	0.38	2.48	Intercooling	(Tobiesen et al., 2007)
6	MDEA – 10 PZ – 30	0.26	0.72	2.49	Solvent split flow	(Dubois & Thomas, 2018)
7	MDEA – 10 PZ – 30	0.17	0.79	2.66	Rich solvent recycle	(Dubois & Thomas, 2018)
8	MDEA – 30 PZ – 20	0.1	0.5631	2.74	N/A	(Zhao et al., 2017)
9	MDEA – 10 PZ – 30	0.171	0.783	2.75	N/A	(Dubois & Thomas, 2018)
10	MDEA – 18.5-18.27 PZ – 21.5-21.23	0.179-0.151	0.721-0.718	2.688-2.764	N/A	(Hosseini-Ardali et al., 2020)
11	MDEA – 35 PZ – 15	0.11	0.31	2.92	N/A	(Tobiesen et al., 2007)
12	MDEA – 10 PZ – 30	0.27	0.74	2.95	Lean vapor compression	(Dubois & Thomas, 2018)
13	MDEA – 45 PZ – 5	0.04	0.5631	3.40	N/A	(Zhao et al., 2017)
14	MDEA – 28-20 PZ – 2-10	0.053-0.077	0.56-0.78	3.0-4.1	N/A	(Khan et al., 2017)

A comprehensive review has been conducted to improve PCC technology to reduce efficiency penalty, energy demand and total cost. The term "Efficiency Penalty" refers to the loss in power generation caused by installing a CCS system in a power plant. It is calculated by subtracting the plant's thermodynamic performance after installing CCS from its performance before installation (Goto et al., 2013). A different process modification was introduced to enhance capturing efficiency and results in reducing the energy demand for regeneration stage (Sachde & Rochelle, 2014). The effect of process modifications to reduce energy demand is represented in **Figure 10** showing examples from the literature about the effect of process modifications on the energy demand for the three selected solvents. Although the system used MDEA promoted by PZ required the highest energy for regeneration before modification, the energy demand reached 2.24 MJ/kg_{CO2} after process modification that is considered the lowest energy amount compared with other solvents. Indeed, adding absorber intercooling is one of the major modifications in the process that

facilitates controlling the absorber temperature, which enhances the process efficiency and reduces energy demand.

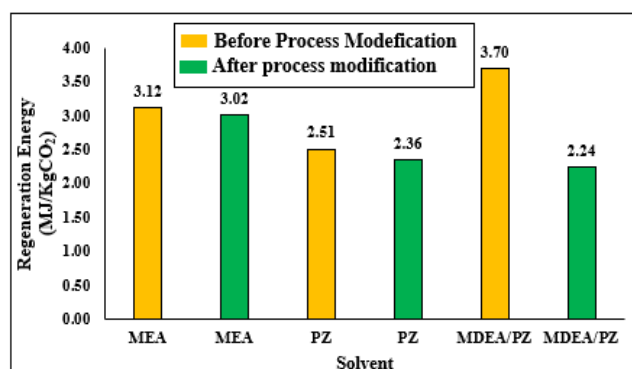


Figure 10 Regeneration energy for the highly utilized solvents before and after process modification.

The synergistic effect of MDEA/PZ is preferable due to the resistance to thermal and oxidative degradation in both absorber and stripper columns, higher stability once compared with MEA, increased absorption capacity, and lower reboiler duty (Closmann et al., 2009; Zhao et al., 2017).

2.4 Environmental assessment studies of solvent-based PCC for CO₂ capture

2.4.1 Solvent-based PCC process notorious emissions to the environment

The emission from the degradation of amines (solvent) is one of the challenging tasks pertaining to absorption PCC process, the triggers of the issues such as the amount and type of solvent, the flue gas composition and others are fully investigated (Supap et al., 2011). Although the solvent degradation causes reduction in the amount of the solvent, the degradation products are documented as highly toxic to human and environment with a possibility of death in specific cases (Rey et al., 2013). Nitrosamines and nitramines are the most dangerous degradation products due to their toxicity such as carcinogenicity and mutagenicity (Magee, 1971). The laboratory tests carried out by Dai et al. to assess the potential of selected solvents (MEA, MDEA, and PZ) to generate nitrosamines when mixed with a combination of Nitric Oxide (NO) and Nitrogen Dioxide (NO₂) revealed that PZ produced the highest amount of nitrosamines, followed by MDEA and MEA (Dai et al., 2012). According to Liang et al.

degradation of amine and the formation of emissions need further investigations (Liang et al., 2015). Furthermore, to enhance capture efficiency using minimum energy demand should not be studied solely without emphasis on the solvent degradation impact. Heat Stable Salts (HSS) is a side-product of PCC processes which is formed by the reaction of amine and its degradation products, once the HSS forms the solvent could not be generated in the stripper (Supap et al., 2011). The HSS include acetate, formate, thiosulfate, sulfate, thiocyanate, oxalate, butyrate, and propionates (Dai et al., 2012). The formation of HSS is to be avoided by preventing or reducing the solvent degradation as minimum as possible (Liang et al., 2015), solvent degradation in PCC are described in **Figure 11**. Solvent degradation hazards are mainly carcinogenicity, mutagenicity, and severe toxicity. It has been shown in the **Figure 11** that toxic compounds are formed in the i) Absorber column wherein it affects the efficiency of CO₂ capture and increases the solvent demand, ii) Stripper column: the formed compounds react with solvent to form HSS, which in turn disturbs and reduces the process efficiency, iii) Washed water: at the top of the absorber column water is located to wash the clean flue gas, the degradation compounds are carried by water to the sewage system and iv) finally the degradation compounds escape to the atmosphere either through leakage or with the flue gas.

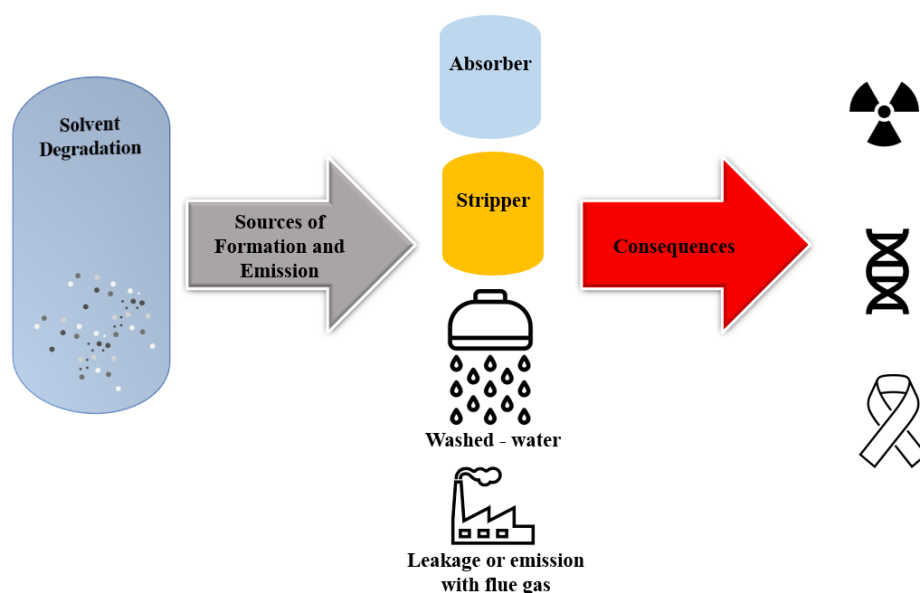


Figure 11 Power plant showing regions of solvent degradation and impact of hazardous compounds on life

2.4.2 Life cycle assessment studies related to solvent-based PCC process

The high reliance on fossil fuel for electricity generation inherently leads to emissions of CO₂ to the atmosphere. However, increasing the integration of renewable energy to generate electricity that has no CO₂ emissions will need time. Meanwhile, solvent-based PCC process appears to be one of the mitigation solutions to reduce the CO₂ emissions (von der Assen et al., 2013). Although the process consumed high energy for solvent regeneration and CO₂ compression that is provided mainly from burning of fossil fuel, still the PCC process shows high efficiency to capture CO₂ from the flue gas (Boot-Handford et al., 2014). However, in addition to providing energy for PCC, the supply of water, as well as the waste and emissions generated by the process, must be further studied to ensure of its environmental sustainability. This reflects the importance of conducting an environmental assessment of the PCC process to ensure it is favorable and sustainable from an environmental perspective (Peters et al., 2011).

Life Cycle Assessment (LCA) serves as an assessment method and analytical tool for evaluating the environmental impacts associated with the flow of materials into and out of the environment for specific processes like Carbon Capture Utilization (CCU). It encompasses everything from raw material extraction, through product production and processing, to recycling or final waste disposal (from cradle to grave) (Curran, 2012). The concept was originated by Chapman et al., who estimated the energy budget in the UK and predicted its future implications (Chapman et al., 1974).

The LCA system includes input and output known as economic and elementary flow (von der Assen et al., 2014). The economic flow refers to the inputs and outputs associated with the process, while the elementary flow represents the exchanges that occur between the process and the environment throughout its life cycle. This flow encompasses various environmental impacts, such as toxic emissions and resource depletion (Curran, 2012).

While the need for LCA is crucial before implementing industrial processes, the complexity and lack of reliable data for conducting LCA have limited the published assessments of PCC process units (Griffiths et al., 2013; Jaramillo et al., 2009; Khoo et al., 2011; Müller et al., 2020).

According to the International Organisation for Standardisation (ISO) the ISO 14040 series of standard, the LCA can be perform in four steps 1) define the goal and scope of the system, 2) data collection through life

cycle inventory, 3) assess the life cycle impact, and 4) interpret the obtained results (Curran, 2006). The tutorial review provides a structures approach to enhance the use of LCA by presenting a checklist of action items that covers all the steps involved in LCA process (von der Assen et al., 2014). Starting with defining the purpose of LCA and the system boundaries, followed by data collection and environmental impact computation, to end up with the interpretation and sensitivity analysis of the results, are covered by the entire process.

Commercial software is available to be used for conducting LCA such as GaBi (Pauer et al., 2020; Petrescu et al., 2017; Spatari et al., 2001), SimaPro (Clarens et al., 2016; Dong et al., 2022; Gładysz & Ziębik, 2015), and Integrated Environmental Control Model (IECM) (Matin & Flanagan, 2022; Pehnt & Henkel, 2009; Schakel et al., 2014). The usage of software ensures the user is following the right LCA methodology since the guidance is provided by the software at the same time.

The literature survey shows that the LCA studies are related to measuring the emissions from power plant with or without PCC such as, Global Warming Potential (GWP), Energy Use (EU), Water Use (WU), and Energy Return on Investment (EROI) (Forster et al., 2007; Hertwich et al., 2008; Koornneef et al., 2008; Spath et al., 1999), following ReCiPe midpoint impact assessment as shown in **Table 12** or following CML 2001 method that developed by the center of environmental science at the University of Leiden (Lacy et al., 2015). Other studies are related to the fuel and power plant system (Odeh & Cockerill, 2008; Pehnt & Henkel, 2009; Singh et al., 2011), and the LCA methodologies have been applied (Finnveden & Moberg, 2005; Suh & Huppes, 2005; Viebahn et al., 2007). Indeed, the LCA studies are not solely to identify the environmental impact factors of different process stages, but they also help policy makers to make proper decisions to achieve clean energy utilization and production (Gaete-Morales et al., 2019).

Table 12 ReCiPe midpoint impact assessment (Kotagodahetti et al., 2021).

Climate Change	Damage to Eco-system
• Climate change • Ozon depletion	• Acidification • Eutrophication (freshwater, marine) • Ecotoxicity (terrestrial, freshwater , marine)
Resource depletion	Human health
• Water depletion	• Human toxicity • Ionizing radiation

• Metal depletion • Fossil depletion	• Photochemical oxidant formation • Particulate matter	• Land occupation (agriculture, urban)
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The summary of the LCA studies related to power plant and CC is listed in **Table 13**, which shows that the standard PCC process includes the four units (absorber, stripper, heat exchanger, and boiler). However, the LCA of the PCC was considered as one unit without further investigations of the impact of each specific equipment such as the absorber and stripper independently. Furthermore, the GWP was identified as the common result obtained across all studies, while the other results such as WU, EU and acidification varied. Most of the data used for LCA was obtained from secondary sources rather than from simulation and modeling (Dones et al., 2007).

Table 13 Summary of the LCA studies of the power plant equipped with CC.

No.	Data source	LCA purpose	LCA Boundary	PCC system	Results ^a	Ref.
1	Scientific literature	Test if Biofuel is the best power plant fuel for 100 year	From Fuel production up to CO ₂ compression – considered PCC as one unit	Standard using MEA	• GWP • EU • WU • EROI	(Bennett et al., 2021)
2	Scientific literature	Global warming reduction from power plant with and without PCC	From coal production up to carbon capture – consider PCC as one unit	Standard using MEA	• GWP • EU	(Branco et al., 2013)
3	Ecoinvent database V2.2	Compare the environmental impact of power plant without and with two carbon capture options	From cradle to gate (fuel extraction up to CO ₂ compression)	Standard using MEA	• GWP • ODP • HTP • PHOF • PMF • TAP • WU • FD	(Clarens et al., 2016)
4	EBSILON 13.02 software	Compare the environmental impact of power plant without and with carbon capture using	From coal production up to carbon captured storage or utilization	Standard using MEA	• GWP • ODP • TAP • EP • HTP • MDP	(Dong et al., 2022)

		either electricity or solar energy for solvent regeneration for 30-years				
5	- Public data - Ecoinvent database	Compare the environmental impact of Taaan power plant without and with PCC	From power plant up to carbon capture	Standard using MEA	- GWP - ODP - TAP - EP - HTP - MDP - WU	(Jung et al., 2022)
6	Ecoinvent database	Compare the environmental impact of three different power plants in the Netherlands without and with PCC	From coal extraction up to carbon capture and storage	Standard using MEA	- GWP - ADP - ODP - HTP - TEP - AP - EP - MAE	(Koornneef et al., 2008)
7	Ecoinvent database	Identify the GWP of power plant with and without PCC	From natural gas production up to EOR of the capture CO ₂	Standard using MEA	GWP	(Lacy et al., 2015)
8	Ecoinvent database	Compare the environmental impact of power plant without and with two carbon capture options	From power plant up to carbon capture	Standard using MEA with different concentration	- GWP - WU - ODP - AP - EP	(Matin & Flanagan, 2022)
9	Ecoinvent database	Compare the environmental impact of power plant without and with carbon capture in China	From coal extraction up to carbon capture and storage	Standard using MEA	- GWP - AP - HTP - EP - ODP - RDP	(Asante-Okyere et al., 2016)
10	- Literature - Aspen Plus	Compare the environmental impact of power plant without and	From coal extraction up to carbon capture and storage	Standard using MDEA	- GWP - AP - EP - ODP - HTP	(Petrescu et al., 2017)

		with three carbon capture options			• MAE • TEP	
11	• Aspen Plus	Compare the environmental impact of four different fuel for power plant with PCC	Power plant up to carbon capture	Standard using MEA	• GWP • AP • EP • ODP • HTP • MAE • TEP	(Romero-García et al., 2022)
12	• Literature	Compression of environmental emission of using different fuels in both power plant and cement industry with and without PCC	From fuel mining up to carbon capture and storage	Standard using MEA	• GWP	(Volkart et al., 2013)

^a The nomenclatures of the results are as following:

GWP = Global Warming Potential

EU = Energy Utilized

WU = Water Utilized

EROI = Energy Return on Investment

EP = Eutrophication Potential

ODP = Ozone Depletion Potential

HTP = Human Toxicity Potential

PHOF = Photochemical Oxidant Formation

PMF = Particulate Matter Formation

TAP = Terrestrial Acidification Potential

FD = Fossil Depletion

MDP = Metal Depletion Potential

ADP = Abiotic Depletion Potential

MAE = Marine Aquatic Ecotoxicity

A comparison study of LCA for the coal fired power plant with and without PCC process shows that 72% reduction on the GWP for 100-year time horizon is expected once PCC is retrofitted to the power plant although, the energy and water demand are increased. Though, the study recommended that the LCA boundary, the fuel used, and the implemented technology to provide power for the PCC process affect the LCA results (Branco et al., 2013). Another study related to comparison of the environmental impact of CC using solvent-based PCC and CaO looping

methods shows that climate change category was 0.33 kgCO₂/kWh for solvent-based PCC compared to 0.26 kgCO₂/kWh using CaO looping and 0.97 kgCO₂/kWh for power plant without capture. The study concluded that CaO looping could be a future alternative for PCC (Clarens et al., 2016).

The LCA studies of PCC to identify the impact of using bioenergy fuel for power plant compared with fossil fuels is reported (Bennett et al., 2021). Around eight CC technologies were tested, and the study found that IGCC power plant is the best option because of no environmental impact, less costly, and low water demands. A full LCA of the power plant includes the waste formation and the reaction of solvent with CO₂ of PCC process was studied (Koornneef et al., 2008). The study compared three scenarios of power plant, the first represented the operating one, second the newly proposed one, and the third scenario represented the newly proposed, equipped with PCC using MEA as solvent. The study found that the GWP is reduced by 71% and Marine Aquatic Ecotoxicity (MAE) is dropped by 27% due to the equipped PCC. However, the other environmental indicators show an increase due to equipped PCC compared to the original power plant case. Indeed, the increase of the environmental indicators reached the maximum for Human Toxicity Potential (HTP) by 181% and Eutrophication Potential (EP) by 80% compared to the original power plant case.

The previous studies of the LCA of the environmental impact of CC have considered the capture system as one unit without evaluating the contribution of individual components or equipment within the system. Additionally, they haven't proposed specific strategies for CC in general or for the PCC process, aimed at reducing environmental emissions (Kotagodahetti et al., 2021). At the same time, the data pertaining to CC are obtained from literature rather than being imported from modeling or simulation system which causes an increase in uncertainty of the applied data for CC part.

The LCA study carried out by He and coworkers (He et al., 2019) assessed the impact of using CC process in power plants, with the system modeled using Aspen Plus. The simulation results were then applied to the LCA study. However, the study also tested a new proposed CC method, chemical looping combustion where the mining stage was identified as the largest contributor to the environmental impact.

The LCA study by Petrescu (Petrescu et al., 2017) evaluated four different scenarios: a power plant without CC, a plant with PCC using MDEA

without a promoter which creates uncertainty regarding the energy needed for solvent regeneration. The study also examined a power plant with PCC using aqueous ammonia, and a plant using calcium looping. Each scenario was analyzed to compare the environmental impact and efficiency of different CC processes. The data of the PCC part of the study was obtained from Aspen Plus simulation while the LCA was conducted using GaBi software. The study concluded that the GWP is highest once power plant operated without PCC (0.97037 kgCO₂/kWh) and that value dropped to 0.49593 kgCO₂/kWh once power plant is equipped with PCC using MDEA. On the other side, the minimum of GWP (0.4022 kgCO₂/kWh) is obtained for power plant equipped with PCC using calcium looping that is similar to a reported study of Clarens and coworkers (Clarens et al., 2016). The literature review identified that PCC is an optimum option for capturing CO₂ from power plant. However, it found that only three solvent according to their physical and chemical properties, experimental studies, and pilot plant utilization are the best option to capture CO₂. Furthermore, the process has a drawback due to the high energy demand for solvent regeneration. Several studies have investigated the process configuration, and in some cases studied the effect of using blended solvent but it was only for open cycle process. The environmental assessment studies using LCA were limited to PCC using MEA and most of the studies were taken data from secondary sources.

2.5 Summary

The literature review of the PCC process using solvent that is carried out in this chapter found that:

- The PCC process is a potential for reducing the emissions of CO₂ from power plant. The drawbacks of this process include the high energy demand for solvent regeneration and process operation, and the environmental impact of the chemicals used during the process. In addition, the limitation and somehow absence of policies either incentives or adverse on the country level are considered as part of the PCC process challenges.
- Several pilot plants are identified in the literature, and open access experimental data of these pilot plants are used for model simulation and validation. The model simulated using pilot plant data following rate-based model shows reliability and accuracy to be used for investigating a solution to the PCC challenges such as process

configuration, process optimization, new solvent, and environmental impact assessment rather than using equilibrium-based modelling approach.

- At the beginning of the study and investigation, MEA was used as the optimum solvent for the capture of CO₂ in both pilot plants and modelling. At a later stage and due to the MEA constraints identified in the literature, other solvents are investigated such as PZ that shows a high ability to capture CO₂ compared with MEA. In the coming years, synergetic efficiency will be the promising solution to enhance the utilization of solvent-based PCC, specifically MDEA/PZ. In addition, the advanced PCC process is more energy and cost effective compared with the standard PCC process, although, the use of MDEA/PZ was not investigated properly in that system.
- Several studies have shown that the environmental impact resulting from solvent degradation and chemical reactions within the PCC process can be hazardous. The release of harmful emissions from the PCC system is most commonly associated with leaks from the absorber and stripper, emissions in the flue gas exiting the absorber column, or contaminants in the system's effluent water. Indeed, the formation of hazardous emissions is directly affected by the process variables and the system configuration.
- The environmental assessment of the PCC process using the LCA procedure shows the ability to identify the impact. However, the reviewed LCA studies on PCC primarily focus on the use of MEA solvent alone, treating PCC as a single unit within the CCS process without analyzing the environmental impact of each individual PCC unit or equipment. Additionally, most studies concentrate on the standard PCC process, despite evidence that advanced processes demonstrate superior technical and economic performance compared to the standard approach.

Chapter 3

Methodologies and Procedures

Methodologies and Procedures

3.1 Overview

This chapter describes the methodologies used in this thesis that presented in **Figure 12** that contain both qualitative and quantitative methods. It highlights the stages involved in the study according to the set aims and objectives. The study involves 4 stages starting with identifying the optimum CC processes, followed by multi-regression analysis to investigate the highly significant variables, after that HYSYS was used for PCC modeling and simulation to investigate the solvent concentration with least regeneration energy requirements, and finally SimaPro used for LCA for the power plant with and without PCC using either MEA or MDEA/PZ to evaluate the environmental impact of each option.

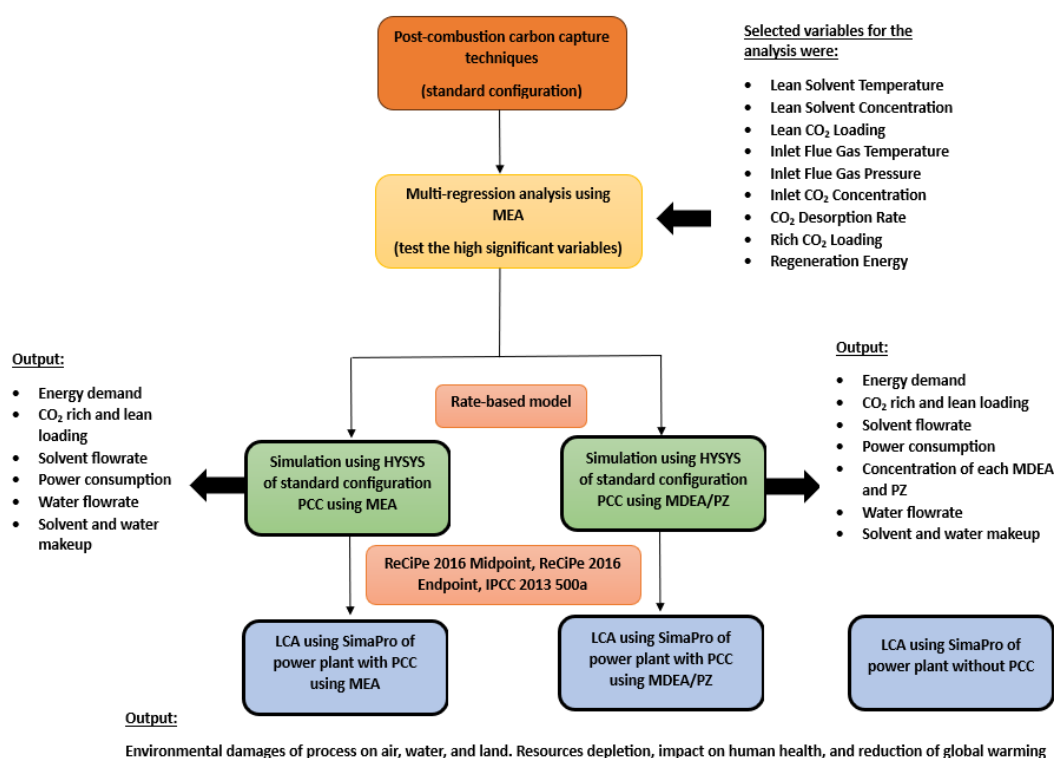


Figure 12 Overview of the methodology used in this thesis

3.2 PCC pilot plant process description

The Commonwealth Scientific and Industrial Research Organisation (CSIRO) constructed a PCC pilot plant in Australia in three different locations to capture CO₂ from coal-fired power plants using chemical absorption process. The PCC in Queensland is flexible for testing different PCC process configurations (Cousins et al., 2011a). The Tarong power plant, is designed according to the standard PCC process (Sultan et al., 2021b; Wang et al., 2015) as shown in **Figure 13**, the process consists of a pretreatment column which needs to remove gaseous pollutants from flue gas and adjust the flue gas temperature. The cooled flue gas is sent to the absorber to capture CO₂ using 30 wt% of MEA. The lean flue gas from the top of the absorber is washed by water to recover any traces of solvent in a separate column. While the rich solvent is sent to the stripper for solvent regeneration and CO₂ separation. The lean solvent (regenerated solvent) is sent back to the absorber in order to reenter into the process (Cousins et al., 2011b; Cousins et al., 2011a). The properties of the PCC pilot technical data are shown in **Table 14** that is designed using a rate-based model, while chemical reactions taking place within the PCC process are shown in **Table 15**.

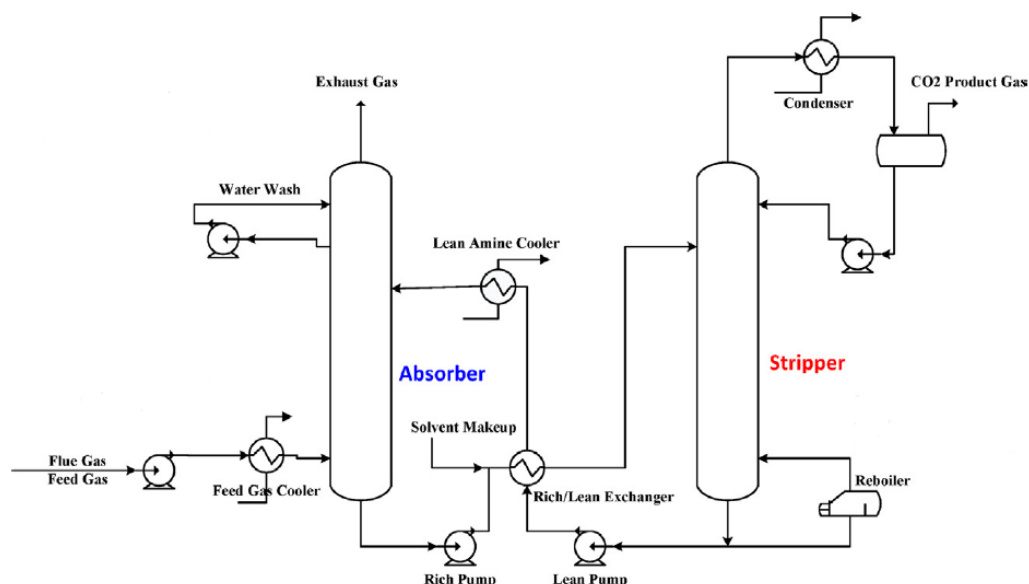


Figure 13 Diagrammatic representation of PCC using chemical absorption (Liang et al., 2015)

Table 14 Technical specifications of the Tarong pilot plant (Cousins et al., 2011a; Li et al., 2016)

Properties	Value
Flue gas	
CO ₂	14% Volume
H ₂ O	8% Volume
N ₂	72% Volume
O ₂	6% Volume
Pressure	1 atm
Temperature	130°C
Absorber	
Solvent (MEA)	30%, Weight
Packing section	4
Packing structure	Sulzer Mellapak M250X
Stripper	
Packing section	2
Packing structure	Sulzer Mellapak M350X

Table 15 Common chemical reactions taking place in PCC process.

S. No	Type	Reaction
1	Equilibrium	$2\text{H}_2\text{O} \rightleftharpoons \text{H}_3\text{O}^+ + \text{OH}^-$
2	Equilibrium	$2\text{H}_2\text{O} + \text{CO}_2 \rightleftharpoons \text{H}_3\text{O}^+ + \text{HCO}_3^-$
3	Equilibrium	$\text{H}_2\text{O} + \text{HCO}_3^- \rightleftharpoons \text{CO}_3^{2-} + \text{H}_3\text{O}^+$
4	Equilibrium	$\text{HOCH}_2\text{CH}_2\text{NH}_3^+ + \text{H}_2\text{O} \rightleftharpoons \text{HOCH}_2\text{CH}_2\text{NH}_2 + \text{H}_3\text{O}^+$
5	Kinetic	$\text{CO}_2 + \text{OH}^- \longrightarrow \text{HCO}_3^-$
6	Kinetic	$\text{HCO}_3^- \longrightarrow \text{CO}_2 + \text{OH}^-$
7	Kinetic	$\text{MEA} + \text{CO}_2 + \text{H}_2\text{O} \longrightarrow \text{MEACOO}^- + \text{H}_3\text{O}^+$
8	Kinetic	$\text{MEACOO}^- + \text{H}_3\text{O}^+ \longrightarrow \text{MEA} + \text{CO}_2 + \text{H}_2\text{O}$

3.3 PCC pilot plant process variables

Evaluation of the significant effects of the process variables is important for identifying the optimum process conditions to achieve the required efficiency. Thus, the expected highly effective process variables to the regeneration energy in the stripper column and the rich CO₂ loading that passes from absorber are listed in **Table 16**. The selected independent variables are lean solvent temperature, lean MEA concentration, lean CO₂

loading, inlet flue gas temperature, inlet gas pressure, inlet CO₂ volume%, and CO₂ desorption rate. While the dependent variables are rich CO₂ loading and regeneration energy. Selected variable and their units are summarized in **Table 16**.

Table 16 Multi-regression dependent and independent variables with respective units.

Variable	Unit
Independent	
lean solvent temperature	°C
lean MEA concentration	wt%
lean CO ₂ loading	mol/mol
inlet flow gas temperature	°C
inlet gas pressure	kPa
inlet CO ₂ volume%	v%
CO ₂ desorption rate	Kg/h
Dependent	
rich CO ₂ loading	mol/mol
regeneration energy	MJ/KgCO ₂

3.4 Multi-regression model description

A plethora of research has investigated the absorption-PCC process model and the ability of the selected solvent to reduce the regeneration energy (Bui et al., 2014; Chikukwa et al., 2012; Llano-Restrepo & Araujo-Lopez, 2015). The selected process's variables for this study are considered in the literature as key variables to find the optimum condition of PCC process (Llano-Restrepo & Araujo-Lopez, 2015). Multi-regression analysis is used to develop a linear mode to investigate the significance of the relationship between the selected process variables using IBM Statistical package for Social Science 25 (SPSS 25). Stepwise regression is applied where variables that have no, or weak significant are excluded. Moreover, the obtained multi-regression equation is validated against the multi-regression assumption before it is used for testing and predicting process variables under different conditions. The model being applied is shown in **Equation 1** and **Equation 2** and **Table 17**, whereas the sample variables are listed in **Appendix 1**. Approximately 22 runs of the performed PCC pilot test are tested.

$$y_1 = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_4 + \beta_5 x_5 + \beta_6 x_6 + \beta_7 x_7 \quad \text{Equation 1}$$

$$y_2 = \beta_8 + \beta_9 x_1 + \beta_{10} x_2 + \beta_{11} x_3 + \beta_{12} x_4 + \beta_{13} x_5 + \beta_{14} x_6 + \beta_{15} x_7 \quad \text{Equation 2}$$

Table 17 Nomenclature of Sample variables.

Variable	Nomenclature
Independent	
lean solvent temperature	x_1
lean MEA concentration	x_2
lean CO ₂ loading	x_3
inlet flow gas temperature	x_4
inlet gas pressure	x_5
inlet CO ₂ volume%	x_6
CO ₂ desorption rate	x_7
Dependent	
rich CO ₂ loading	y_1
regeneration energy	y_2
Coefficient	
regression coefficients	$\beta_0, \beta_1, \beta_2, \beta_3, \beta_4, \beta_5, \beta_6, \beta_7, \beta_8, \beta_9, \beta_{10}, \beta_{11}, \beta_{12}, \beta_{13}, \beta_{14}, \beta_{15}$

3.5 Model development for PCC process

The model of PCC process was developed in Aspen HYSYS V12.1 using acid gas property package that follows rate-based model approach. The model includes column hydrodynamics and detailed component mass transfer calculations between liquid and vapor phase. Furthermore, the model includes calculation of properties of amines being utilized, CO₂ lean and rich, and the makeup unit (water and solvent makeup). Rate-based model has been used in an extensive research and it exhibits reliable and accurate performance for process simulation (Zhang et al., 2009). The liquid phase of the model was calculated using the Electrolyte Non-Random Two-Liquid (ENRTL) thermodynamic model and vapor phase properties was calculated using Peng-Robinson's equation, Equation 3, Equation 4, Equation 5. The model developed through Aspen HYSYS is shown in **Figure 14**.

In this work, the data is taken from the pilot plant constructed by the Commonwealth Scientific and Industrial Research Organisation (CSIRO) in Australia (Cousins et al., 2011a). The reasons of using the CSIRO pilot plant is the PCC process designed according to the standard model that captured CO₂ from flue gas emitted from Tarong power plant (Cousins et al., 2011b) and the comprehensive experimental data that simplified the validation of PCC process model (Li et al., 2016). The technical specifications of PCC pilot plant are listed in **Table 18**.

$$P = \frac{RT}{V-b} - \frac{a(T)}{V(V+b)+b(V-b)} \quad \text{Equation 3}$$

$$a = 0.4278 \frac{R^2 T_c^{2.5}}{P_c} \quad \text{Equation 4}$$

$$b = 0.0867 \frac{R T_c}{P_c} \quad \text{Equation 5}$$

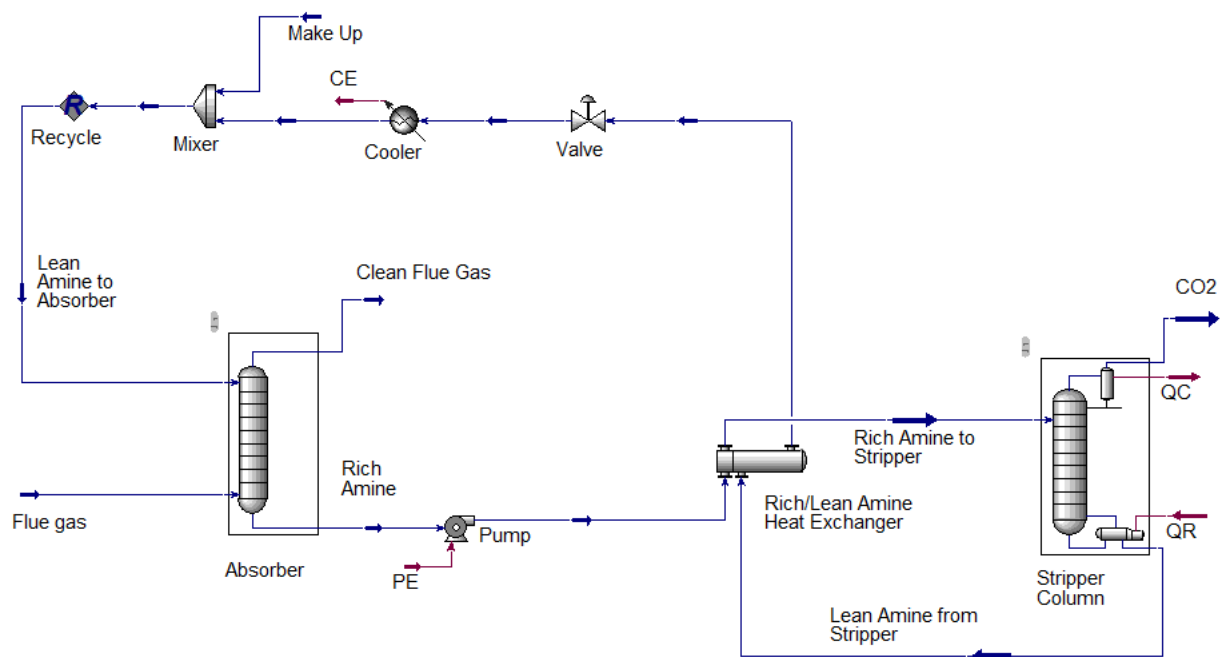


Figure 14 The developed HYSYS model of PCC process for this study

Table 18 Technical specifications of the Tarong Pilot Plant

Properties	Value
Flue gas	
CO ₂	14% Volume
H ₂ O	8% Volume
N ₂	72% Volume
O ₂	6% Volume
Pressure	1 atm
Temperature	130°C
Absorber	
Solvent (MEA)	30%, Weight
Packing section	4
Packing structure	Sulzer Mellapak M250X
Stripper	
Packing section	2
Packing structure	Sulzer Mellapak M350X

The assumptions used in the model includes:

- Steady-state model.
- No accumulation in the bulk gas as well as the liquid and gas films.
- The feed to the system was only the flue gas as listed in **Table 18** and the MEA solvent.
- The mass and energy balance for both liquid and gas phase are formed in one-dimensional.
- The flow of flue gas and solvent follows counter-current mode.

The developed PCC process consists of the absorber, pumps, heat exchangers, stripper, and compressor as depicted in **Figure 14**. Flue gas emitted from burning fossil fuel is sent to the absorption unit for CO₂ capture in the range 80-90% in the flue gas by the selected solvent which leaves the column as rich solution. The absorber column is designed to enhance the mass transfer between the flue gas and the selected solvent using structured packing. The temperature of CO₂ rich solvent is adjusted using rich/lean heat exchanger prior to being sent to the stripper column. There in the column, the solvent is separated from the captured CO₂ and sent back to the absorber column as lean solution, while the captured CO₂ is compressed and stored in the geological storage area. The efficiency of the PCC process depends on the performance of chemical reactions take place in the absorber and the stripper columns as listed for MEA in **Table**

15. Absorption of CO₂ is taken place through acid/base reaction where the solvent reacts with CO₂ according to the reactions presented in **Table 15**.

3.6 Model simulation using MDEA/PZ for PCC process

The PCC model using MDEA/PZ blended solvents was developed in Aspen HYSYS V12.1 following the same model for MEA as described in Section 4.2. where the assumptions used in the model includes:

- Steady-state model.
- No accumulation in the bulk gas as well as the liquid and gas films.
- The feed to the system were only the flue gas as listed in **Table 18**, PZ, and MDEA solvents.
- The mass and energy balance for both liquid phase and gas phase is one-dimensional.
- The flow of flue gas and solvent follows counter-current mode.

The sensitivity analysis carried on for the findings from the simulation model using MDEA/PZ under standard conditions of the PCC process. The purpose of the sensitivity analysis for the simulated model is to assess the variation of the key input variables within the selected specific range those are:

- Solvent concentration: 40 – 50 wt%
- Solvent flow rate: 60 – 172 Kgmole/h
- CO₂ lean loading: 0.13 – 0.28 mole_{CO2}/mole_{amin}
- CO₂ rich loading: 0.23 – 0.41 mole_{CO2}/mole_{amin}

The key input variables are examined toward the performance of energy demand for solvent regeneration, solvent degradation rate, viscosity, and process efficiency. Hence, the proposed statistical model for the study is as shown in Equation 6 that studies the correlation between the solvent concentration, solvent flow rate, CO₂ lean loading, and CO₂ rich loading against the energy demand for solvent regeneration.

$$y = f(\text{solvent concentration, solvent flow rate, CO}_2 \text{ lean loading, CO}_2 \text{ rich loading})$$

Equation 6

where, y is the energy demand for solvent regeneration.

The second proposed statistical model is as shown in Equation 7 that studies the synergistic effects of solvents toward solvent viscosity that left the absorber and the stripper.

$$\text{solvent viscosity} = f(\text{solvent concentration, solvent flowrate, regeneration energy})$$

Equation 7

The dependent variables for the proposed statistical model are energy demand and solvent's viscosity, while the independent variables are the solvent's concentration, its flow rate, CO₂ lean loading, and CO₂ rich loading. The statistical assumption maintaining the required process efficiency for CC from flue gas.

The efficiency of the PCC process depends on the performance of the chemical reactions taken place in the absorber and the stripper columns as listed in **Table 19** for MDEA/PZ. Absorption of CO₂ is taken place through acid base reaction where each solvent (MDEA and PZ) reacts with CO₂ according to the reactions presented in **Table 19**. Chemical reactions 1-3 represent the dissociation of water, formation of carbonate, and formation of bicarbonate, respectively. At the same time, PZ acts as activator for MDEA related reaction (reactions 4 and 5). Bicarbonate is formed as a result of MDEA reaction with CO₂ that enhances the absorption and regeneration performance, while carbamates is formed as a result of PZ reaction with CO₂ that enhances the CO₂ absorption rate.

The closed loop model of the PCC system is considered the best option that provides accurate and reliable results for investigating technical and economic process aspects. However, the reported studies of closed loop system were either related solely to PZ (Otitoju et al., 2021) or MEA (Akula et al., 2021). However, the closed loop system modeling and simulation using MDEA/PZ has not been reported in literature (Closmann et al., 2009; Khan et al., 2017).

Table 19 Chemical reactions in PCC process (Hosseini-Ardali et al., 2020).

No	Category	Reaction
1	Water related	$2\text{H}_2\text{O} \rightleftharpoons \text{H}_3\text{O}^+ + \text{OH}^-$
2		$\text{H}_2\text{O} + \text{HCO}_3^- \rightleftharpoons \text{H}_3\text{O}^+ + \text{CO}_3^{2-}$
3		$\text{CO}_2 + \text{OH}^- \rightleftharpoons \text{HCO}_3^-$
4		$\text{MDEA} + \text{H}_3\text{O}^+ \rightleftharpoons \text{MDEAH}^+ + \text{H}_2\text{O}$

5	MDEA Related	$\text{MDEA} + \text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{MDEAH}^+ + \text{HCO}_3^-$
6	PZ related	$\text{PZ} + \text{H}_3\text{O}^+ \rightleftharpoons \text{PZH}^+ + \text{H}_2\text{O}$
7		$\text{PZCOOH} + \text{H}_2\text{O} \rightleftharpoons \text{PZCOO}^- + \text{H}_3\text{O}^+$
8		$\text{PZ} + \text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{PZCOO}^- + \text{H}_3\text{O}^+$
9		$\text{PZCOO}^- + \text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{PZCOOH} + \text{HCO}_3^-$
10		$\text{PZCOO}^- + \text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{PZ}(\text{COO})_2^{2-} + \text{H}_3\text{O}^+$
11		$\text{PZH}_2^{2+} + \text{H}_2\text{O} \rightleftharpoons \text{PZH}^+ + \text{H}_3\text{O}^+$

The validated HYSYS model of the closed PCC process (**Figure 14**) is used to investigate the synergistic effect of MDEA/PZ on the regeneration energy, the power needed to drive the pump and the cooler, the viscosity of the blended solvents, the individual concentration of the solvents, and the lean solvent flow rate. The other process variables such as the pressure and temperature of absorber and stripper, the flue gas flow rate, and the CO₂ vol% in the flue gas were kept constant for the purpose of the study.

3.7 PCC system formulation using blended solvent

As discussed earlier, an Aspen HYSYS model is simulated based on PCC pilot plant constructed by Commonwealth Scientific and Industrial Research Organisation in Australia (Cousins et al., 2011a) to capture CO₂ from 350 MW coal-fired Tarong power plant. The developed PCC model aimed to investigate the optimum concentration of MDEA/PZ to capture CO₂ that required the minimum energy for solvent regeneration compared to the benchmark MEA at 88.5% CC rate. The total energy demand, water flow rate, solvent flow rate, and other process variables were estimated by Aspen HYSYS V12.1 using acid gas property package that follows rate-based model approach. The energy and flow rate related findings from HYSYS model are illustrated in **Table 20**, representing the best MDEA/PZ technical option compared to the benchmark MEA. For further details related to the coal-fired Tarong power plant refer to the reported works (Cousins et al., 2011b; Cousins et al., 2011a; Li et al., 2016).

Table 20 Finding from HYSYS simulation for PCC model.

Solvent	Concentration (wt%)	Solvent flow rate (kg/hr)	Total power demand (KW)	Total water flowrate (kg/hr)	Regeneration energy (MJ/KgCO ₂)	Makeup solvent (kg/hr)	Makeup water (kg/hr)
MEA	27.84	475.55	152.622	1136.61	4.003	0.055	20.92
MDEA/PZ	43/7	1131.24/ 184.16	107.985	1215.16	2.53	0	0

3.8 Description of LCA methodology

The methodology of LCA is used to investigate the environmental impact for a product and process throughout the life cycle of three scenarios including coal-fired power plant without PCC, coal-fired power plant with PCC using MEA, and coal-fired power with PCC using MDEA/PZ. The LCA involves four stages including goal and scope, life cycle inventories, life cycle impact assessment, and results interpretation (Clarens et al., 2016; ISO, 2006; Miller et al., 2011).

The SimaPro 9.3.0.2 professional software is utilized following methods of ReCiPe 2016 Midpoint (H) (Goedkoop et al., 2009), ReCiPe 2016 Endpoint (H) (Huijbregts et al., 2017), and IPCC 2013 500a (IPCC, 2013) to conduct the LCA of the mentioned three scenarios. An optimal amount of 1 MWh electricity generated is selected as a reference for the resources consumed and pollutants discharged.

3.8.1 Goals and Scope

The goal of the LCA of this study is to evaluate the environmental impact of utilizing PCC unit with MEA for one scenario and blended solvent containing MDEA/PZ for another scenario compared to resources consumption and pollutants emissions from coal-fired power plant without PCC that is producing 350 MWh of electricity. This resulted in providing a framework as a guidance for decision-makers to enable enhance energy efficiency and follow a clean and sustainable energy production strategy. The LCA system boundary is illustrated in **Table 20**, **Figure 15** and **Figure 16** for the system without PCC and with PCC, respectively. Furthermore, the LCA study boundary “cradle-to-gate” approach (Müller et al., 2020) is used for this study, that illustrated in **Figure 17**. The other stages of LCA include construction and transportation, were excluded from the study because of the similarity of the power plant and to enable evaluation of the PCC process and identification of the environmental effect of synergistic solvents specifically compared to the solo solvent. The intention of the

study was to find out the possibility of the blended solvent (MDEA/PZ) to be replaced by MEA for capturing CO₂ from a power plant.

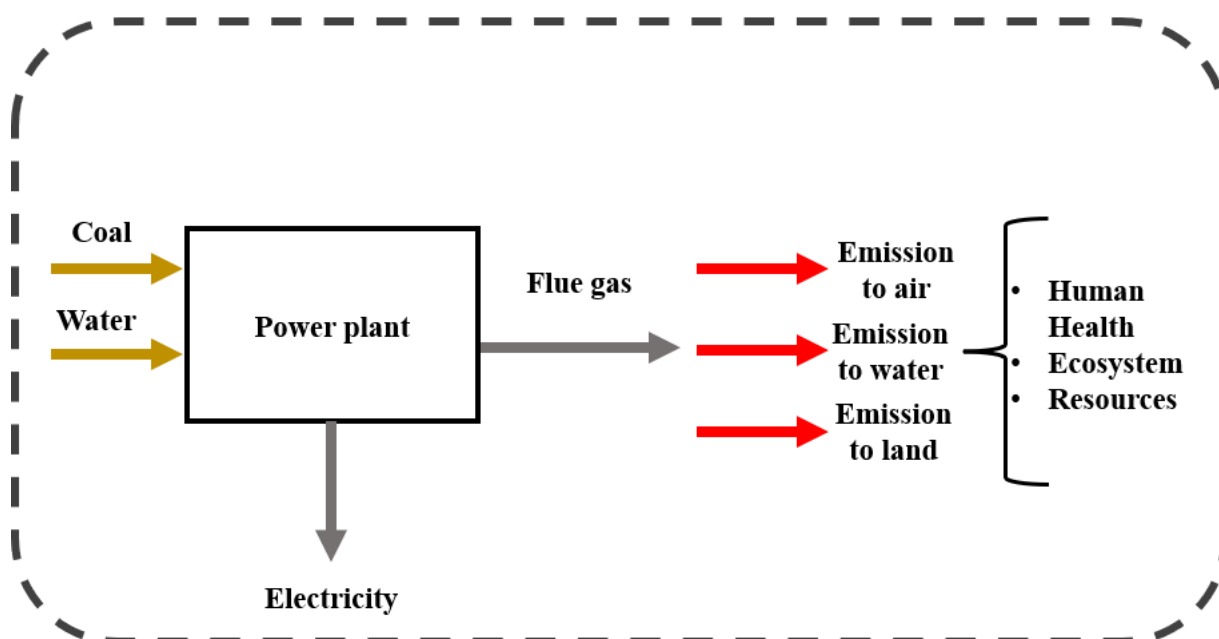


Figure 15 Flowchart of this work LCA boundary without PCC.

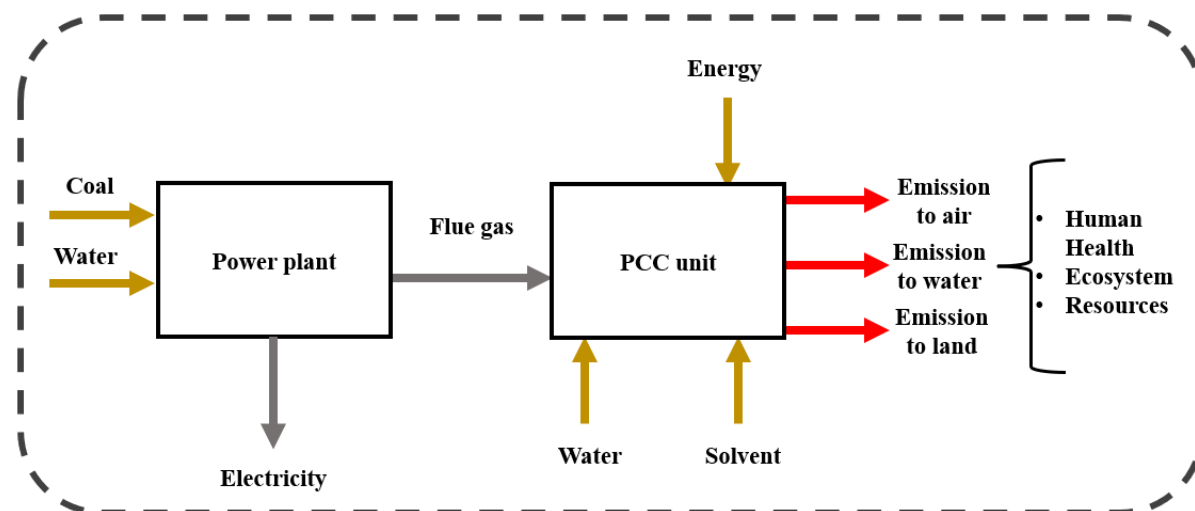


Figure 16 Flowchart of LCA boundary with PCC, applied in this study.

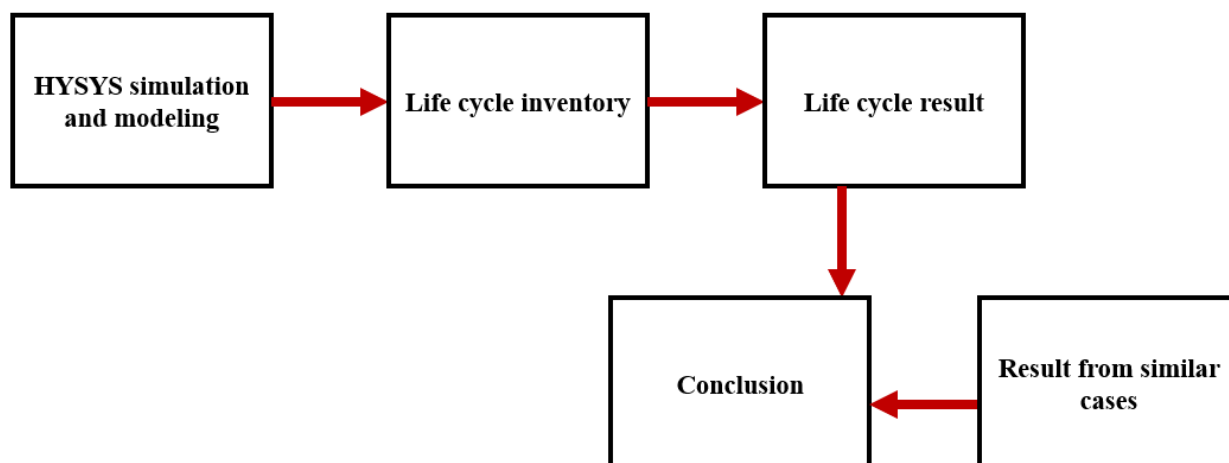


Figure 17 System processing and analysis steps.

3.8.2 Inventory and information

The purpose of (LCI) part is to collect the data related to the process input and output of raw materials, energy demand and emissions within the system boundary for a functional unit of 1 MWh of electricity generated in the power plant with a life time of 30 years (Rasheed et al., 2021). The data for the LCA is taken from the simulated HYSYS model where the data related to coal process are taken from Ecoinvent v3.5 database of SimaPro v.9.3 that is considered as a reliable data source (Wernet et al., 2016). The life cycle assumptions for the three scenarios are presented in **Table 21**.

Table 21 LCA assumption and calculations for the three scenarios.

Process	Unit	Value	Reference
Coal-fired power plant			
Total fuel amount	t/h	135,450	Calculated Calculated (Petrescu et al., 2017)
Total water demand	m ³ /t	0.4186	
Electricity for coal mining operation and preparation	MJ/t	0.79	
CO ₂ emissions	Kg/h	84.8075	(Li et al., 2016)
H ₂ O in the flue gas	Kg/h	11.1796	(Li et al., 2016)
O ₂ in the flue gas	Kg/h	52.2038	(Li et al., 2016)
N ₂ in the flue gas	Kg/h	340.4091	(Li et al., 2016)
PCC System			
MEA consumption	Kg/tCO ₂	5.6	Output of HYSYS model
MDEA consumption	Kg/tCO ₂	13.34	
PZ consumption	Kg/tCO ₂	2.17	

3.8.3 Life cycle impact assessment of the overall process

The LCIA was carried out using software SimaPro v.9.3 following the methodology of ReCiPe 2016 Midpoint (H) (Goedkoop et al., 2009), ReCiPe 2016 Endpoint (H) (Huijbregts et al., 2017), and IPCC 2013 500a (IPCC, 2013). Indeed, LCIA is the significant step of LCA where the environmental emissions and resources are translated to categories or scores under two mainstream ways either at midpoint or endpoint level (Goedkoop et al., 2009; Hauschild & Huijbregts, 2015). In this study, the obtained data was used to evaluate the system impact against 3 endpoint environmental indicators including natural resources, human health, and ecosystem. They are linked to 7 midpoint impact categories, which show high discrepancy among the selected scenarios, including global warming potential (GWP), water consumption potential (WCP), ozone formation terrestrial ecosystem potential (OFTEP), Freshwater Ecotoxicity potential (FWEP), marine ecotoxicity potential (MEP), ozone formation human health potential (OFHHP), and terrestrial ecotoxicity potential (TEP) (Curran, 2012).

3.8.3.1 ReCiPe 2016 methodology

The ReCiPe 2016 is a methodology for assessing the environmental impact of the midpoint and endpoint that measures the problem and damage, respectively. It follows the Hierarchist (H) perspective that is based on the most common policy principles (Huijbregts et al., 2017). At the midpoint level the following categories are addressed:

1. Climate change: it measures the GWP due to carbon feedstock and substances that contribute to global warming over specific time horizon. The unit is year/kg CO₂ equivalent (IPCC, 2013).
2. Water use: it measures the water consumption potential of freshwater in m³. The category gives indication about the sustainability level of the applied practice (Döll & Siebert, 2002).
3. Photochemical ozone formation, terrestrial ecosystems: it measures the ozone formation terrestrial ecosystem potential in the unit of year/kg NO_x. The factor indicates the rate of ozone formation due to the emissions of particles such as NO and NO₂ that form ozone in the atmosphere (Van Zelm et al., 2016).
4. Freshwater eutrophication: it measures the Freshwater Ecotoxicity potential due to the effluent from the source that contains

environmental persistence hazardous chemicals to the freshwater resources (Helmes et al., 2012).

5. Marine eutrophication: it measures the marine ecotoxicity potential by measuring the effluent from the source that contains environmental persistence hazardous chemicals to the marine ecosystem (van Zelm et al., 2009).
6. Photochemical ozone formation, human health: it measures the ozone formation human health potential. The factor indicates the rate of ozone formation in the atmospheric layer due to the emissions of particles such as NO and NO₂ and the impact level on human health (Van Zelm et al., 2016).
7. Human toxicity and ecotoxicity: it measures the terrestrial ecotoxicity potential due to the emissions of highly hazardous chemicals that accumulate in the human food chain and cause toxicity. The unit is kg/year 1,4-dichlorobenzene (1,4-DCB) (van Zelm et al., 2009).

The characteristics of endpoint that assess the damage are listed as below:

1. Human health: it measures the impact of the emissions on human health and the disabilities across a numbers of years (De Schryver et al., 2011).
2. Ecosystem: it measures the losses of species and degradation of the ecosystem as a result of emissions from the source across a numbers of years (Huijbregts et al., 2017).
3. Resources scarcity: it measures the possibility of natural resources depletion such as fossil fuels, and future resources production due to the source regular practice across a numbers of years (de Schryver et al., 2008).

Chapter 4

Results, Discussion, and Recommendations

Results, Discussion, and Recommendations

4.1 Variables highly significant on CO₂ rich loading

The multi-regression output using the stepwise method shows that only three independent variables out of the selected seven are highly correlated and significant to the rich CO₂ loading, as shown in **Table 22**. The first model shows a correlation between the CO₂ desorption rate and the rich CO₂ loading. The correlation increases to reach the maximum of 0.837 when the standard error reached the minimum as shown in Model 3 that identified the highly correlated variables. The highly significant variables are the CO₂ desorption rate, lean CO₂ loading, and lean solvent temperature. Indeed, the rich CO₂ loading is the output from the absorption process that should indicate the efficiency of captured CO₂. According to the multi-regression result, the optimum capture is achieved once the optimum values of the three highly correlated independent variables are obtained.

Table 22 Model Summary of the rich CO₂ loading using multi-regression model.

Model	R	R Square	Adjusted square	R	Std. error of the estimation
1	0.627 ^a	0.393	0.363		0.01118
2	0.751 ^b	0.564	0.518		0.00972
3	0.837 ^c	0.700	0.650		0.00828

^aPredictors: (Constant), CO₂ desorption rate (x_7)

^bPredictors: (Constant), CO₂ desorption rate (x_7), Lean CO₂ loading (x_3)

^cPredictors: (Constant), CO₂ desorption rate (x_7), Lean CO₂ loading (x_3), Lean temperature (x_1)

Equation 8 to be used for prediction. The equation shows that the three independent variables are directly proportional to the CO₂ rich loading which reflect the same findings, shown in **Table 22**. Thus, to increase the capture efficiency, the change to the lean solvent temperature should not be higher than 0.002 between the two runs of the PCC pilot test and could be attributed to the increase of the absorber column temperature due to reaction in the absorption column. The higher temperature within the absorber column enhances the reversible reaction that leads towards separation of CO₂ from MEA. On the other side, a higher lean CO₂ loading helps to increase the capturing efficiency due to the increase in partial pressure of CO₂ within the absorber column. Although a higher lean CO₂ loading is the result of low desorption process, increases the CO₂ partial

pressure thus enhances the capture efficiency. Hence, to achieve the maximum rich CO₂ loading, the maximum desorption rate must be obtained under a minimum value of lean solvent temperature and a minimum amount of lean CO₂ loading. According to Equation 8, statistically, lean CO₂ loading (x_3) is a strong significant variable to the rich CO₂ loading compared to other variables.

$$y_1 = 0.304 + 0.002 * x_1 + 0.202 * x_3 + 0.001 * x_7 \quad \text{Equation 8}$$

4.2 Variables highly significant for regeneration energy

Stepwise method is used to find out the highly correlated independent variable toward regeneration energy. The multi-regression output shows that three independent variables are highly correlated with the regenerating energy, **Table 23**. The system identified around three models, where the first one shows the correlation between lean MEA concentration and regenerating energy. The second model shows correlation with inlet CO₂ amount and MEA concentration. The correlation between the variables increases to reach the maximum of 0.943 (strong positive correlation) with a minimum standard error for Model 3 (**Table 23**). The highly correlated variables are lean MEA concentration, inlet CO₂ amount, and lean CO₂ loading. In fact, regeneration energy is required to separate CO₂ from MEA, where MEA is recirculated from stripper to the absorber to capture the next CO₂ feed. The amount of energy regeneration needs to be reduced to avoid reduction of the efficiency of the power plant, where power plant is provided with the energy required for regeneration stage. Accordingly, the optimum minimum energy is required once the best amount of the three highly correlated independent variables is obtained.

Table 23 Summary of the regeneration energy using multi-regression model.

Model	R	R Square	Adjusted square	R	Std. error of the estimation
1	0.638 ^a	0.407	0.377		0.14570
2	0.877 ^b	0.768	0.744		0.09339
3	0.943 ^c	0.890	0.871		0.06622

^aPredictors: (Constant), Lean MEA concentration (x_2)

^bPredictors: (Constant), Lean MEA concentration (x_2), Inlet CO₂ Volume (x_6)

^cPredictors: (Constant), Lean MEA concentration (x_2), Inlet CO₂ Volume (x_6), Lean CO₂ loading (x_3)

The relationship between dependent and independent variables is obtained from the multi-regression equation as shown in equation 9. The equation shows that there is a direct proportional relationship between the regeneration energy and the two independent variables, lean CO₂ loading and inlet CO₂ amount, while there is indirect proportional relationship between the regeneration energy and the lean MEA concentration. Hence, to achieve the minimum regeneration energy, the amount of solvent in the feed should be the optimum, which could be attributed to the required regeneration energy once high amount of solvent is added due to the chemical reactions taken place within the process. The reaction needs to be more in the forward direction towards CO₂ capture rather than desorption and that is achieved once enough solvent is available to capture the maximum amount of CO₂. Although increasing the amount of CO₂ in the flue gases requires higher energy for solvent regenerating, the inlet amount of CO₂ needs to be high to avoid drops in the pressure within the absorber column and reduces the efficiency of the PCC process. Therefore, low CO₂ amount, low lean CO₂ value, and high MEA concentration must be used to achieve high capture efficiency using the minimum regeneration energy. With reference to Equation 9, the amount of solvent and its related chemical reactions are statistically strong significant variables to the regeneration energy compared to other variables. Hence, investigation of a new solvent or testing the synergistic effect could contribute to further reduction of the energy required for solvent regeneration.

$$y_2 = 4.144 - 0.06 * x_2 + 0.103 * x_6 + 2.248 * x_3 \quad \text{Equation 9}$$

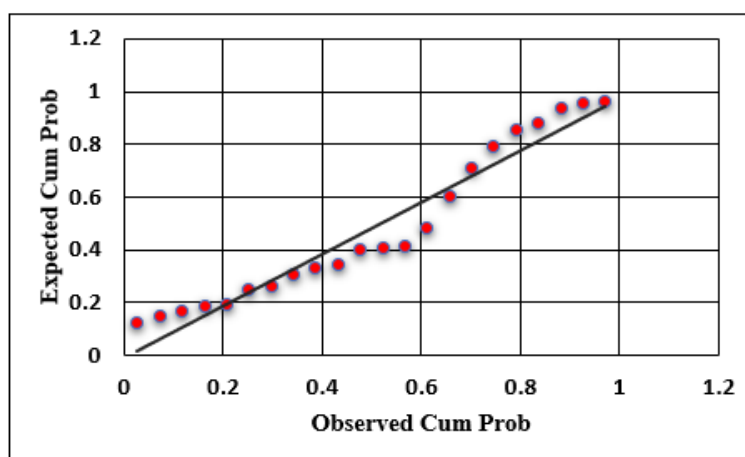
4.3 Multi-regression equation validation

4.3.1 Validation against rich CO₂ loading

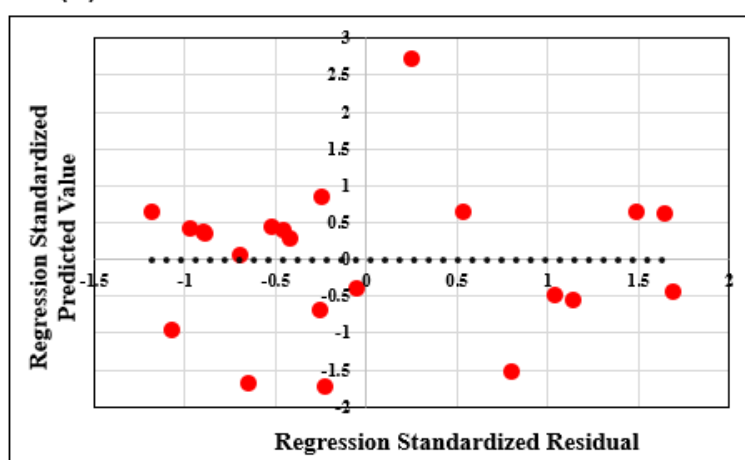
The multi-regression equation for rich CO₂ loading (Equation 8) is validated against the three multi-regression assumptions those are (i) the linearity validation of the correlation between the independent and dependent variables, (ii) the variation in the residuals is same between current and predicted value of the dependent variable which followed normal probability distribution, and (iii) the independent variables are not correlated.

The residual plot shows a relationship between the current dependent variables and the predicted dependent using the regression equation. The

best regression line should pass through the center of the data using scatter plot as shown in **Figure 18a**. It shows that a good amount of data is available above the line and a good number is available below the line which reflects the positive and negative distribution of values. In addition, **Figure 18b** is associated with residuals and shows the random distribution of positive and negative values, which must be centered by the horizontal zero line. The findings in **Figure 18** clearly reveal that the linearity assumption is accurate.



(a)

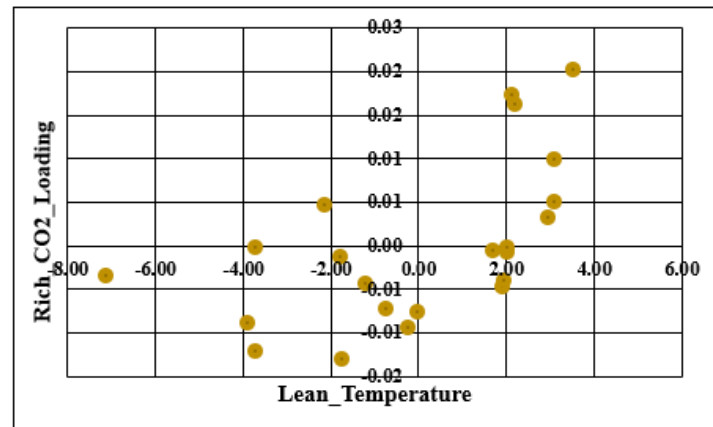


(b)

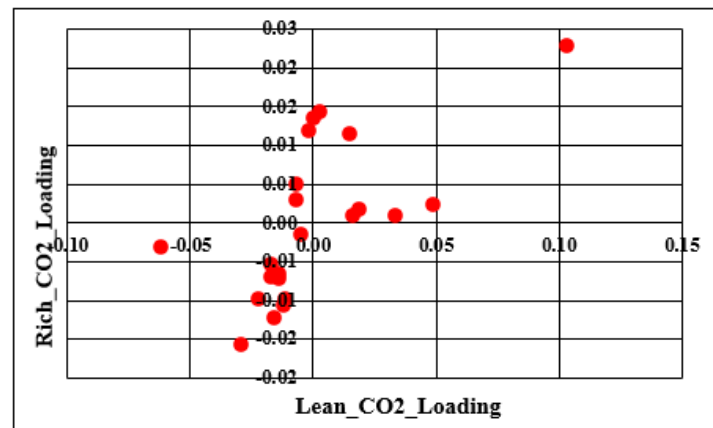
Figure 18 Multi-regression prediction and current testing (CO_2 rich loading) including (a) P-P plot regression standardized residual that shows the difference between the current probability and predicted probability and (b) residual graph that shows the difference between the current and predicted value of CO_2 rich loading.

The linear relationship between dependent and independent variables must be tested to validate the second assumption that examines the ability of Equation 8 to be used for prediction purposes. A graph is drawn for the three independent against the dependent variables as shown in **Figure 23**; the lean temperature correlation with rich CO_2 (**Figure 19a**), lean CO_2

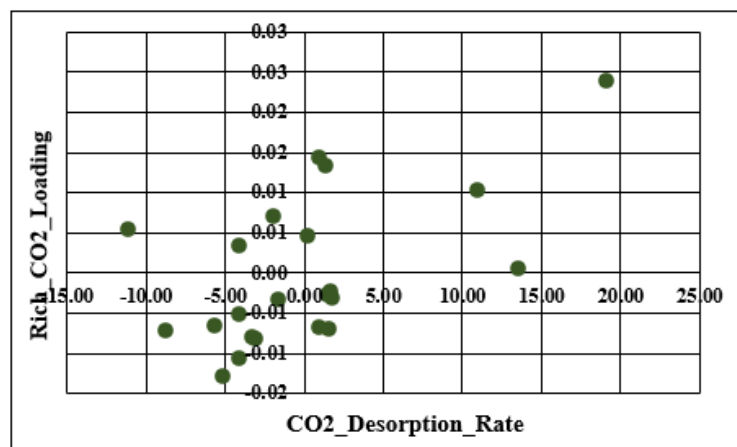
loading (Figure 19b), and the CO₂ desorption rate (Figure 23c). It is evident that each independent variable has a positive strong correlation with dependent variable, hence, the findings validate the second assumption of the multi-regression equation and reflect the accuracy and reliability of the obtained equation.



(a)



(b)



(c)

Figure 19 Correlation of dependent variable and individual independent variables (CO₂ rich loading) including (a) correlation of lean temperature with rich

CO₂ loading, (b) correlation of lean CO₂ loading with rich CO₂ loading and (c) correlation of CO₂ desorption rate with rich CO₂ loading

Typical multicollinearity is used to test the correlation between the independent variables to ensure that the predicted dependent variable is accurate and has no effect on the reliability of the hypothesis test. The Variance Inflation Factor (VIF) calculated by using Equation 10 is used to test the correlation between the independent variables, where R_j^2 values are obtained through SPSS for each independent variable.

$$VIF = \frac{1}{1-R_j^2} \quad \text{Equation 10}$$

The estimated values of VIF are 1.212, 1.035, and 1.213 for the lean solvent temperature, the lean CO₂ loading, and the desorption rate, respectively. The accepted value of VIF for no correlation between the independent variables must be less than 10. Since the estimated VIF values for the three independent values are less than 10 it clearly shows that the obtained regression equation is reliable and can be used for predicting target scenarios.

4.3.2 Validation of equations against solvent regeneration energy

Following the same procedure the multi-regression equation is validated against the CO₂ rich loading. The relationship between current regeneration energy and the predicted one (residual graph) is shown in **Figure 20a**. the regression line crosses the data where some points are above and below the line which indicates the positive and negative distribution of the data, reflecting the best regression line. At the same time, **Figure 20b** shows that the trendline is the zero horizontal line where part of the data is distributed over and below the line. Therefore, the findings in **Figure 20** are consistent with the linearity assumption.

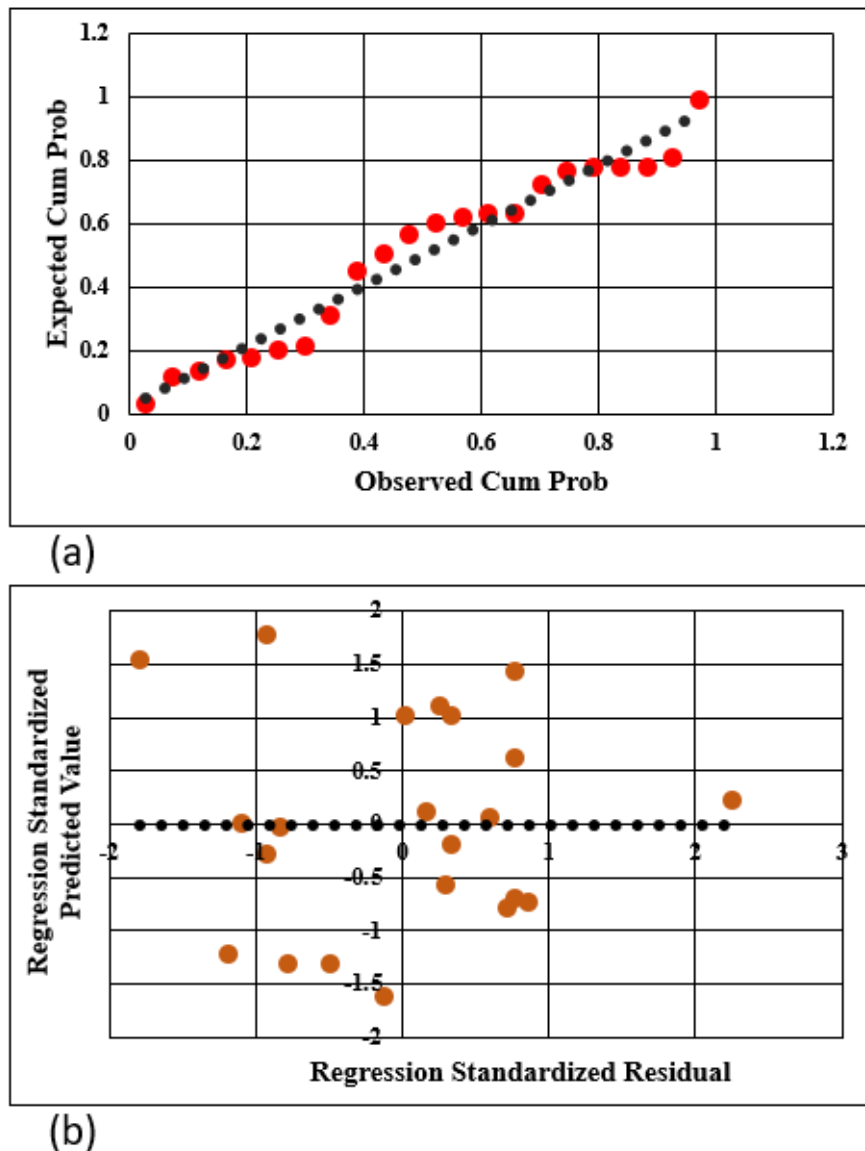
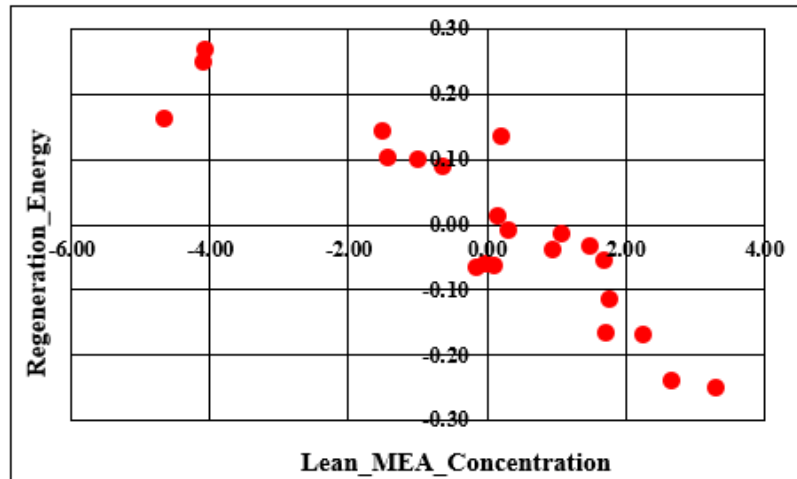
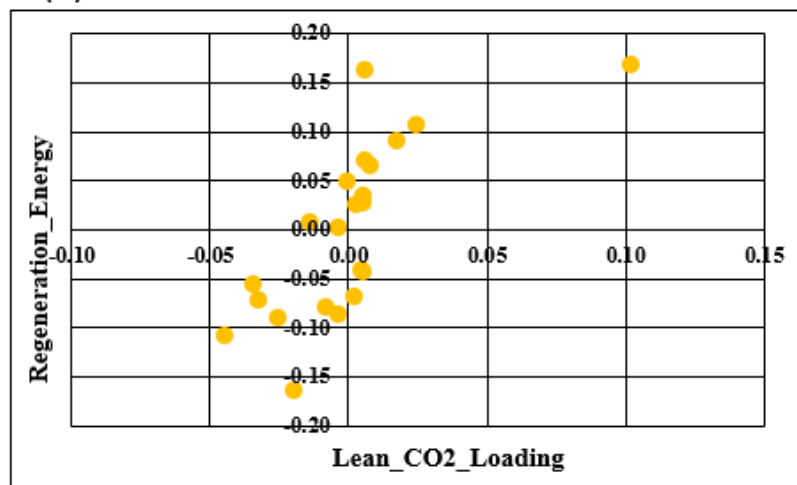


Figure 20 Multi-regression prediction and current testing (Regeneration energy) including (a) P-P plot regression standardized residual that shows the difference between the current probability and predicted probability and (b) residual graph that shows the difference between the current and predicted value of regeneration energy.

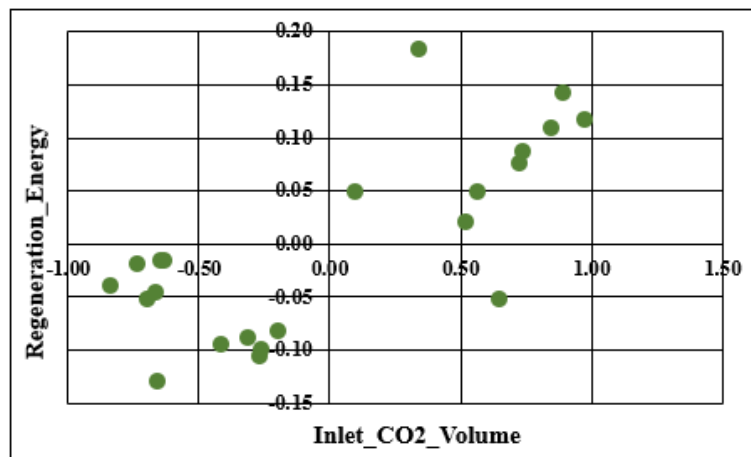
The correlation obtained in **Equation 9** needs to be validated to ensure that a linear relationship occurs between dependent variables and each independent variable. Hence, **Figure 21a** shows the linearity between lean MEA concentration and the regeneration energy, **Figure 21b** shows the linearity between lean CO_2 loading and regeneration energy, **Figure 21c** shows the linearity between inlet CO_2 amount and the regeneration energy. Thus, the scatters represented in **Figure 21** supports the second assumption of the multi-regression equation validation.



(a)



(b)



(c)

Figure 21 Correlation between dependent and independent variables (Regeneration energy) including (a) correlation of the lean MEA concentration with regeneration energy, (b) correlation of the lean CO₂ loading with regeneration energy and (c) correlation of the inlet CO₂ volume with regeneration energy.

The VIF values for the independent variables are estimated using equation 10, it can be found that VIF values are 1.496, 1.772, and 1.315 for the lean MEA concentration, inlet CO₂ amount, and the lean CO₂ loading, respectively. The estimated value of VIF is less than 10, therefore, it supports the reliability of the obtained regression equation for predicting scenarios.

4.4 Reliability of multi-regression equations

The validation results of the multi-regression equations for CO₂ rich loading and regeneration energy are up to the mark and it is assumed that the equations are appropriate to be used for prediction purposes. These equations enable saving efforts and cost. Testing of the multi-regression equations against trustworthy sources is needed to identify the limitation of the equations, if any, and to investigate the accuracy of predicting values. The references selected for equations' testing purpose are listed in **Table 24**. It depicts the values of CO₂ rich loading and regeneration energy from each reference according to the values of the independent variables identified in their works. In addition, the PCC process (standard or modified) is identified.

Table 24 Comparison of dependent and independent variables obtained from different sources.

Sr. No.	independent variables ^a					CO ₂ rich loading ^b	Regen Energy ^c	PCC model ^d	y_1^e	y_2^e	Ref.
	x_1	x_2	x_3	x_6	x_7						
1	30	30	0.31	12	90	0.5	2.727	Modified	0.517	4.277	Frimpong et al. 2019
2	40	30	0.36	16	90	0.49	3.351	Modified	0.547	4.801	Frimpong et al. 2019
3	32.8	30	0.23	8.8	90	0.6	3.8	Standard	0.506	3.767	Mostafavi et al. 2021
4	40	30	0.219	11	90	0.441	2.649	Modified	0.518	3.969	Oh et al. 2020
5	40	30	0.219	11	90	0.441	3.873	Standard	0.518	3.969	Oh et al. 2020
6	40	30	0.2	14.88	90	0.51	3.57	Modified	0.514	4.326	Oh et al. 2018
7	40	30	0.24	13.17	90	0.48	5.76	Standard	0.522	4.240	Bravo et al. 2021

^aThe independent variables are defined in **Table 24** those selected as expected to be highly effects on the PCC process, where x_4 and x_5 ; inlet flow gas temperature and the pressure of the input gas are excluded from the analysis because they show no correlation with the dependent variables according to the multi-regression analysis.

^bThe CO₂ rich loading values are as given in the listed reference.

^cThe regeneration energy is the amount of energy in MJ/Kg_{CO2} required for separating captured CO₂ from MEA to be used again for CO₂ captured and the values are regenerated from the listed references.

^dThe PCC model is either the standard one that consists of absorber, stripper, heat exchanger, and reboiler, while the modified one is developed by the researcher to reduce regeneration energy such as by adding another stripper (Frimpong et al., 2019) or by adding cold solvent split and stripper overhead compression (Oh et al., 2020). The standard model is the one used for multi-regression analysis.

^e The values of y_1 and y_2 are estimated from multi-regression equations used in this work where y_1 is the predicted value of CO₂ rich loading and y_2 is the predicted value of regeneration energy

The predicted values of the CO₂ rich loading using the required variables are plotted against the actual value of CO₂ rich loading, **Figure 25**, where most of the actual values of CO₂ rich loading are closed to the predicted values (Bravo et al., 2021; Frimpong et al., 2019; Oh et al., 2018). However, the difference between the values approaches the maximum for Mostafavi et al. and Oh et al. (Mostafavi et al., 2021; Oh et al., 2020) which is attributed to increase in the lean CO₂ loading and the high temperature of the lean solvent. Indeed, the increase in lean solvent temperature is controlled by intercooling, that is introduced to the middle of the absorber column, where addition of that item is not detected by the regression equation. The comparison (**Figure 22**) gives an indication about the validity and reliability of the multi-regression equation to predict the CO₂ rich loading for standard PCC conditions.

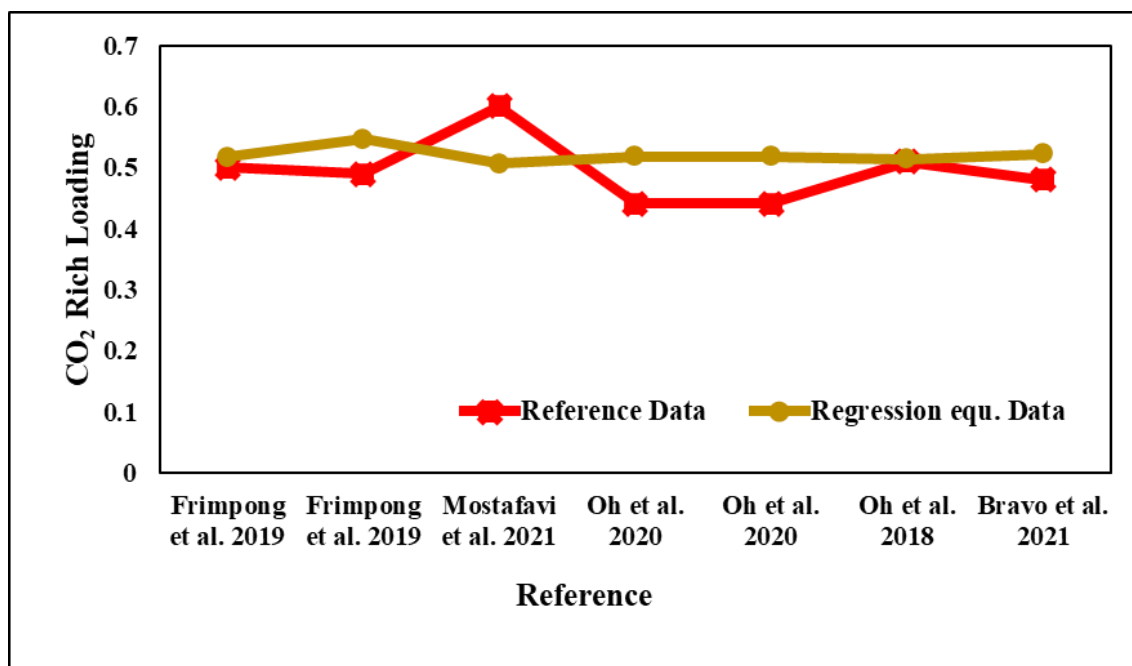


Figure 22 Comparison between literature reports and regression equation (developed in this study) for CO₂ rich loading values.

The predicted values of the regeneration energy are marginally different from the actual regeneration energy for some references, as shown in **Figure 23**. The works of Mostafavi et al. and Oh et al. (Mostafavi et al., 2021; Oh et al., 2020; Oh et al., 2018) show that the actual regeneration energy and predicted regeneration energy values are approximately same, where the PCC model of these works follow the standard one. Contrary to this, approximately 36% reduction in the actual regeneration energy compared to the predicted value is obtained when equation is applied to the Frimpong et al. work. Indeed, that is attributed to the modification done where a developed PCC model is utilized using two strippers. The use of two strippers reduced the load on the reboiler to provide the required energy for regenerating the solvent (Frimpong et al., 2019). However, the regeneration energy of the work reported by Frimpong et al. increases from 2.727 to 3.351 MJ/Kg_{CO2} due to increased lean CO₂ loading and inlet CO₂ amount to the absorber. It reflects the same for the predicted value where the regeneration energy is increased from 4.28 to 4.80 MJ/Kg_{CO2}.

The developed model of Oh et al. (Oh et al., 2020) shows a reduction by 33% of the regeneration energy compared to the predicted value of regeneration energy. The PCC process is configured by lean vapor compression, cold solvent split, and stripper overhead compression. These modifications reduce the pressure of the stripper to provide the required energy for solvent regeneration.

The work of Bravo et al. shows increases by 36% in the actual regeneration energy compared to the predicted value, according to the authors the amount of regeneration energy should be close to 4.2 MJ/Kg_{CO2}, that is similar to the predicted value, while the value increases due to heating and evaporation of the added water (Bravo et al., 2021).

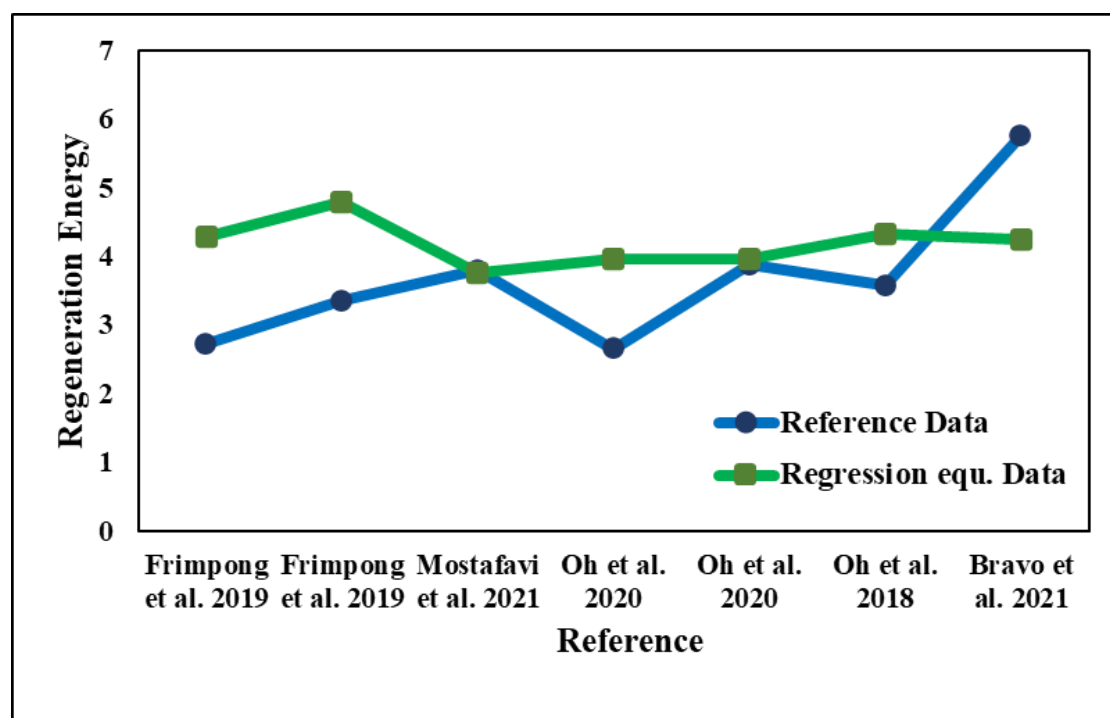


Figure 23 Comparison between reported works and regression equation for the amount of regeneration energy.

Testing of the developed multi-regression equation shows the reliability and accuracy of the equations to be used for predicting the CO₂ rich loading and regeneration energy once the standard PCC model is used. In addition, the developed multi-regression equation can be used as benchmark to compare the effect of process modification to the CO₂ rich loading and regeneration energy. In addition, the multi-regression equation are obtained once one solvent is used following the standard model that is preferable in the industry due to economic factors (Brandl et al., 2021). However, with reference to the obtained results, solvent has a strong effect on regeneration energy. It encourages further investigations of the synergistic effect to reduce regeneration energy value and to enhance capture efficiency following standard PCC model, achieving this target will certainly enhance the utilization of PCC process.

4.5 HYSYS Model validation for PCC process

The performance of the rate-based model described in Chapter 3 is validated using six experiments reported by Li *et al.*, (Li et al., 2016). The detailed of the selected cases are illustrated in Table 25.

The model predictions of CO₂ rich loading, regeneration energy, and CO₂ absorption rate of the PCC process were compared to the experimental data for the six considered cases. The absorption rate is determined using the expression in Equation 11.

$$\text{CO}_2 \text{ absorption rate} = m_1 - m_2 \quad \text{Equation 11}$$

Where m_1 is the flow rate of CO₂ in the flue gas and m_2 is the flow rate of CO₂ in the outlet gas. The CO₂ rich loading is calculated using Equation 12.

$$\text{CO}_2 \text{ rich loading} = \frac{[\text{CO}_2] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] + [\text{MEACOO}^-]}{[\text{MEA}] + [\text{MEA}^+] + [\text{MEACOO}^-]} \quad \text{Equation 12}$$

The results of the rate-based model predictions against the experimental data are shown in Table 26 for CO₂ rich loading, regeneration energy, and CO₂ absorption rate. In each case, the model predictions obtained from HYSYS agree with the experimental data. The percentage errors between the experimental data and the model predictions are calculated using Equation 14.

$$\% \text{ error} = \frac{\text{experimental data} - \text{model prediction data}}{\text{experimental data}} \times 100 \quad \text{Equation 13}$$

The deviations between experimental and simulated results are illustrated in Table 27 that show similarities between the experimental and predicted data.

Table 25 Summary of the operation conditions for the MEA-based PCC pilot plant (Li et al., 2016)

Operating conditions	Value
CO ₂ in the flue gas (vol%)	11.9-13.5
Flue gas temperature (°C)	48.1-65.5
MEA concentration (wt%)	24-30.3
Lean amine flow (L/min)	27-33
CO ₂ captured rate (kg/h)	70.6-76.8
Absorber pressure (Kpa)	105.6-107.5
Stripper pressure (Kpa)	177.5-195
Reboiler temperature (°C)	116.4-117.6
Condenser temperature (°C)	25.4-32
Regeneration energy (MJ/kgco ₂)	4.35-4.5
Lean Loading (mol _{CO2} / mol _{MEA})	0.279-0.333

Table 26 Pilot plant experiments used for model validation versus model predictions.

Category	Case No.					
	1	2	3	4	5	6
CO ₂ rich loading Experiment (mol _{CO2} /mol _{MEA})	0.473	0.486	0.494	0.480	0.469	0.471
CO ₂ rich loading Model (mol _{CO2} /mol _{MEA})	0.488	0.502	0.492	0.496	0.500	0.487
Regeneration energy Experiment (MJ/Kgco ₂)	4.48	4.51	4.49	4.45	4.35	4.5
Regeneration energy model (MJ/Kgco ₂)	4.399	4.15	4.31	4.32	4.003	4.5
CO ₂ absorption rate experiment Kg/h	74.9	78.8	76	71	70.6	73.6
CO ₂ absorption rate model Kg/h	76.01	75.74	77	70.46	74.95	76.80

Table 27 Deviations between experimental and simulation results for the 6 experimental cases

No.	Category	% Error in each case ^a					
		1	2	3	4	5	6
1	CO ₂ rich loading	-3.19	-3.33	0.36	-3.33	-6.5	-3.48
2	Regeneration energy	1.81	7.98	4.01	2.92	7.98	0.00
3	CO ₂ absorption rate	-1.48	3.88	-1.32	0.76	-6.16	-4.35

^a The % error is illustrated the variation of the simulation result from the pilot plant test experiments

The model predictions against the experimental data for CO₂ rich loading, regeneration energy, and CO₂ absorption rate are plotted in **Figure 24**, **Figure 25**, and **Figure 26**. The plots show the model predictions follow the experimental trend that indicates a good agreement. In addition, the range of deviation between the model predictions and experimental results did not exceed $\pm 8\%$ which is reasonable. Hence, it can be concluded that the HYSYS model satisfactorily validated the pilot plant experimental results at the same time, the obtained results validate the reliability of the rate-based model to be used for this purpose.

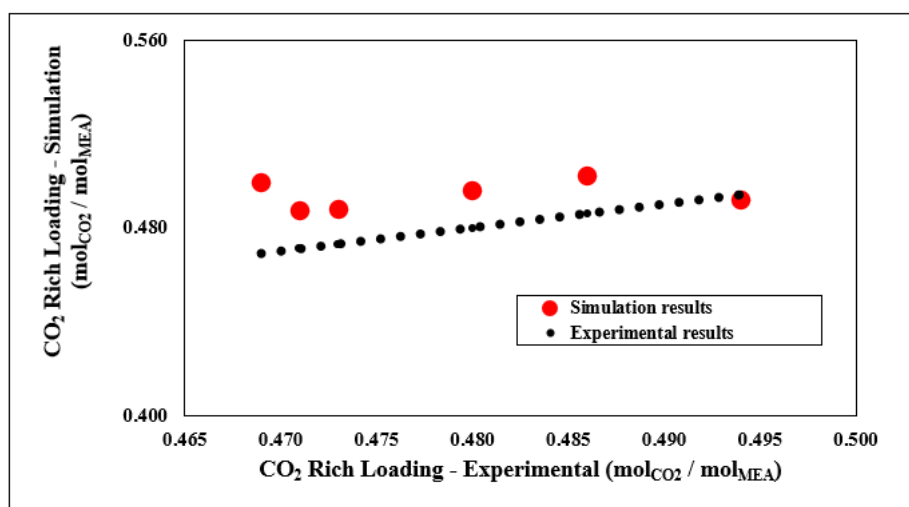


Figure 24 Experimental results versus simulation results for CO₂ rich loading for six experimental cases

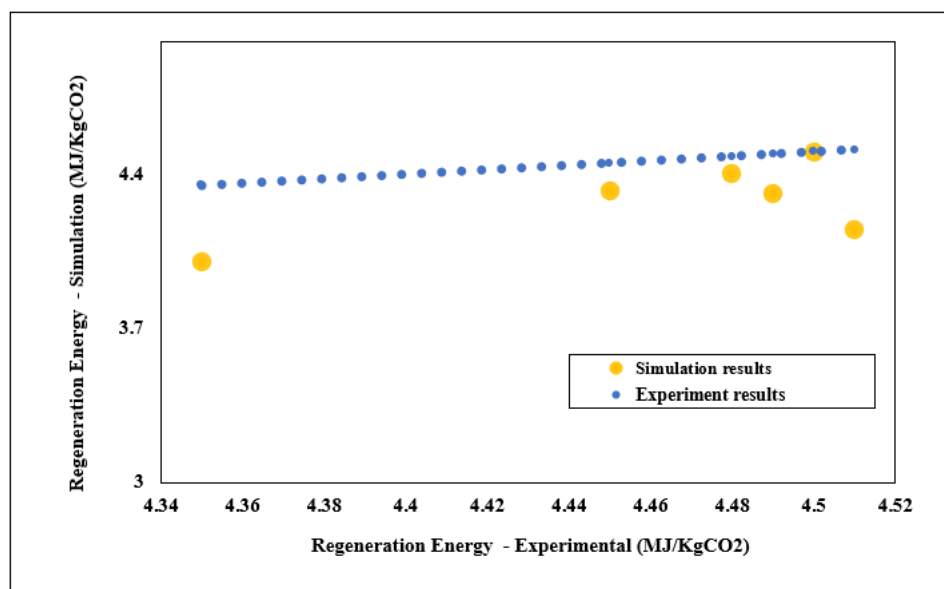


Figure 25 Experimental results versus simulation results for regeneration energy for six experimental cases

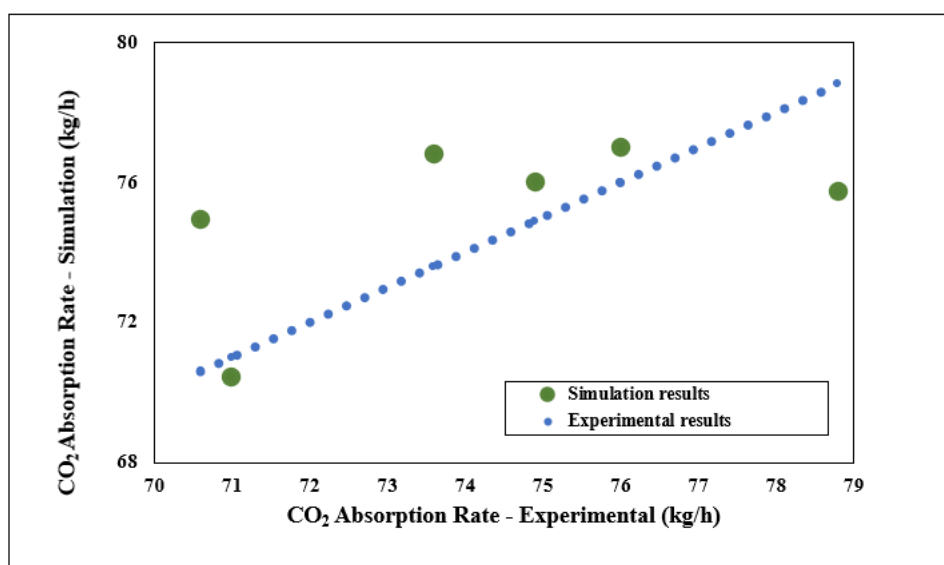


Figure 26 Experimental results versus simulation results for CO₂ absorption rate for the 6 experimental cases

4.6 Analysis of HYSYS Simulation for PCC process

4.6.1 Effect of solvent concentration

The described PCC model in chapter 3 simulated using MDEA/PZ at different total-mass concentrations (40 and 50 wt%, respectively). The 40 and 50 wt% of PZ alone or blended MDEA/PZ are considered the optimum as standard solvent concentration for PCC process in various pilot plant. For example, the separation research program at the University of Texas at Austin (USA) (Chen et al., 2013; Dugas, 2006), CSIRO Tarong where 40 wt% of PZ was used to capture CO₂ (Cousins et al., 2015; Rochelle et al., 2019), and CO₂ separation plant in Durnroht (Austria) (Rabensteiner et al., 2015). Furthermore, the 40 and 50 wt% total concentration of MDEA/PZ is considered the optimum in various process simulation (Mudhasakul et al., 2013; Samanta & Bandyopadhyay, 2011; Zhao et al., 2017). For each process run, the flow rate of CO₂ in the lean solvent was fixed to be 2.1838 kgmol/h, while the individual concentrations of MDEA/PZ were varied to investigate the synergistic effect on solvent flow rate, CO₂ lean loading, CO₂ rich loading, viscosity, solvent and water makeup, power consumed, and regeneration energy.

The solvent flowrate is one of the key variables that affect the absorption efficiency specifically in the absorption column. The data illustrated in Table 28 are the required to achieve 88.5% capture efficiency for different MDEA/PZ concentrations that are considered as part of the economic

range of the design PCC unit (Rao & Rubin, 2006). According to **Table 28**, the solvent flowrate decreases as the concentration of PZ increases for the two cases (40 and 50 wt%) and that is because of the high efficiency of PZ to absorb CO₂ compared to MDEA (Closmann et al., 2009). The solvent flow rate using 50 wt% total concentration of MDEA/PZ is lower than 40 wt% and that maximizes the capture efficiency and reduces the consumption of thermal energy.

The CO₂ lean and rich loading listed in **Table 28** are within the optimum range as identified in the literature (Notz et al., 2012; Otitoju et al., 2021) which is 0.2 for CO₂ lean loading and 0.4-0.5 for CO₂ rich loading. The viscosity listed in **Table 28** was calculated using indexed mixing rules (Horvath, 1985) for the solvent inlet to the absorber and rich solvent left the absorber for the blended solvents Equation 15.

$$\eta_{mix}^{1/3} = \sum_i x_i \eta_i^{1/3} \quad , \quad \text{where } \eta \text{ is the viscosity} \quad \text{Equation 14}$$

The aim from investigating the variation in solvent viscosity is to ensure no operational problems are associated with the high viscosity, particularly that as the PZ concentration increases, the viscosity of the solution increases (**Table 28**).

According to the finding of Yuan et al, (Yuan et al., 2017) the viscosity of the solvent increases rapidly as the concentration of the solvent increases and it requires reducing the column height to avoid reduction in the diffusivity of solution followed by reduction in the capture efficiency. However, **Table 28** shows that the viscosity of the rich solvent (outlet of absorber column) dropped because of elevated temperature in the absorbed column, causing a drop in the viscosity by 40% for the 50 wt% (MDEA/PZ) solvent blend. Hence, the obtained results support the selection of 50 wt% total (MDEA/PZ) concentration due to high process efficiency compared with 40 wt% total (MDEA/PZ) and that is similar to the finding of (Zhao et al., 2017).

Table 28 Comparison of process efficiency variables for different MDEA/PZ concentration.

MDEA/PZ	MDEA/PZ individual wt%	Solvent flow rate (kgmol/h)	CO ₂ lean loading (moleCO ₂ /mole _{amin})	CO ₂ rich loading (moleCO ₂ /mole _{amin})	Lean solvent viscosity (cP)	Rich solvent viscosity (cP)
40 wt%	38 - 2	171.8	0.1326	0.2362	5.746	4.688
	35 - 5	109	0.2022	0.3601	6.876	4.846
	30 - 10	84.5	0.2459	0.4382	7.512	5.310
	28 - 12	80.75	0.2569	0.4572	7.218	5.094
	25 - 15	75.08	0.2689	0.4788	7.109	4.778
	20 - 20	68.5	0.2865	0.5099	6.888	4.080
50 wt%	43 - 7	81.27	0.1877	0.3343	14.45	8.665
	40 - 10	73.56	0.2034	0.3621	15.000	8.890
	35 - 15	65.8	0.2207	0.3927	14.56	8.019
	30 - 20	60.18	0.2345	0.4173	14.39	7.05

4.6.2 Energy performance evaluation of HYSYS Simulation Process

The regeneration energy for MDEA/PZ being identified for each case is illustrated in **Figure 27**. The regeneration energy increases as the amount of PZ increases and that energy could reach a higher level as the individual concentration of PZ exceeds 20 wt% for the two cases 40 wt% and 50 wt% of total concentration, the same finding is reported by Khan et al. (Khan et al., 2017) but for 30 wt% total concentration. This is due to the formation of carbamate in a higher amount as shown in **Table 19** in which carbamate needs higher energy to release CO₂ compared with bicarbonate (Khan et al., 2017). The lowest identified energy demand is 2.69 MJ/kgco₂ and that reached at (35 wt% MDEA / 5 wt% PZ) for 40 wt% and 2.53 MJ/kgco₂ at (43 wt% MDEA / 7 wt% PZ) for 50 wt% as shown in **Figure 27**. However, the solvent flowrate at 35 wt% MDEA / 5 wt% PZ is higher by 32.5% than 43 wt% MDEA / 7 wt% PZ, hence, the optimum individual concentration is 43/7 wt% (MDEA/PZ) for 50 wt% total concentration.

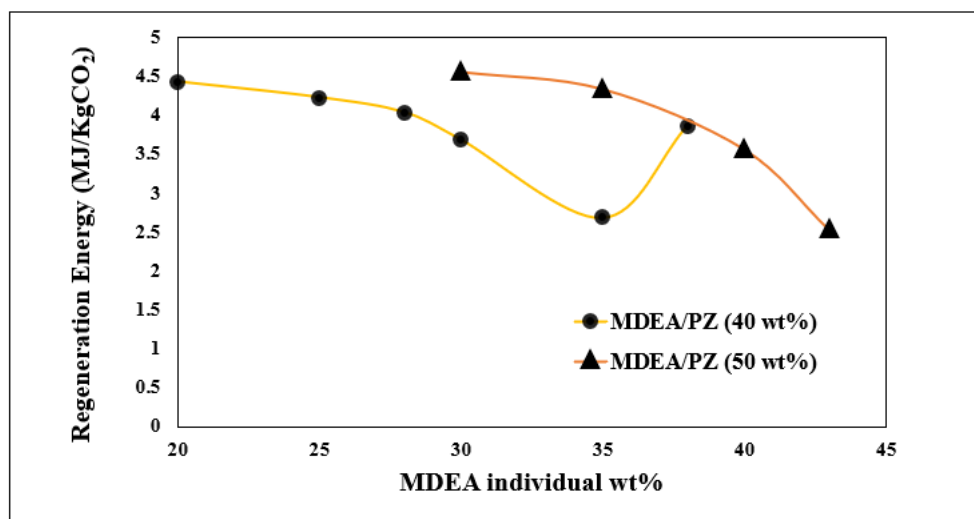


Figure 27 Regeneration energy for different MDEA/PZ concentration (38/2, 35/5, 30/10, 28/12, 25/15, and 20/20 for 40 wt%), (43/7, 40/10, 35/15, and 30/20 for 50 wt%)

The energy performance of different concentration of MDEA/PZ using PCC standard layout evaluated based on the electrical energy to operate the pump, cooler, compressor and the heat duty needed for reboiler is shown in **Table 29**. According to the tabulated results, **Table 29**, reboiler requires the highest energy compared to the other process energy consumers for the two cases 40 and 50 wt%. Nonetheless, the reboiler power reached the minimum (56.24 KW for 40 wt% and 52.49 KW for 50 wt%) for the two cases identified in **Figure 27** which demanded the lowest regeneration energy.

The identified results is considered as an evidence of the synergistic effect of the blended solvent toward reducing the regeneration energy and enhancing process efficiency where optimum concentration needs to be selected. In addition, the power needed for pumping the rich solvent to the stripper column is similar for the studied cases in this work (**Table 29**) and that indicates that the high viscosity of the lean solvent is reduced during the absorption process as a result of increasing the temperature within the column. The high power needed for cooler can be reduced by modifying the design of the lean/rich heat exchanger to enhance the heat transfer between CO₂ rich and lean loading. Indeed, reducing the required power is one of the factors that enhances the utility of PCC. It is because that the needed power for PCC process is provided by electricity generated in the power plant (Abu-Zahra et al., 2007a).

Table 29 Energy consumption of the simulated model.

MDEA/PZ total wt%	MDEA/PZ individual wt%	Pump (KW)	Reboiler (KW)	Compressor (KW)	Cooler (KW)	Total Energy (KW)
40	38 - 2	0.1211	80.35	15.29	67.28	163.0411
	35 - 5	0.07679	56.24	15.02	43.82	115.157
	30 - 10	0.05989	76.850	14.740	64.280	155.930
	28 - 12	0.05701	84.08	15.18	71.25	170.567
	25 - 15	0.05307	88.02	16.53	73.42	178.023
	20 - 20	0.04814	92.29	19.09	72.77	184.198
50	43 - 7	0.06523	52.49	13.97	41.46	107.985
	40 - 10	0.05922	74.29	13.79	62.91	151.049
	35 - 15	0.05311	90.33	15.95	76.04	182.373
	30 - 20	0.04842	94.5	19.48	75	189.028

4.7 Technical performance of synergistic effect on PCC process with reference to MEA

The standard PCC process using 30 wt% MEA is considered the benchmark in which other PCC processes are compared (Canepa et al., 2012; Frimpong et al., 2019; Zhang & Chen, 2013). In this study, the developed standard PCC using MDEA/PZ 40 and 50 wt% are compared to the standard PCC using 30 wt% MEA reported by Li et al. (Li et al., 2016) for the same process specification and the other operation variables. The findings illustrated in **Table 30** reveal that using MDEA/PZ requires less regeneration energy against the benchmark 30 wt% MEA. Although the solvent flow rate using MDEA/PZ is higher than MEA, the regeneration energy reached the minimum of 2.53 MJ/kgCO₂ by 36.8% reduction compared to MEA. In addition, the makeup water and solvent that need to be added to the recycle lean solvent stream are high compared to no makeup requirement for the MDEA/PZ two cases. The addition of the makeup due to solvent losses could indicate either the degradation or evaporation of solvent as part of the clean flue gas where both need further investigation regarding environmental assessment. The CO₂ lean loading and rich loading for the three scenarios give expectation to the ability of blended solvent to achieve the required process efficiency compared to the efficiency of MEA. The required total power for standard PCC as shown in **Table 30** is the highest for MEA and the lowest for MDEA/PZ 50 wt% and it shows the synergistic effect of the solvent to enhance the process efficiency.

Table 30 PCC process specification using MDEA/PZ vs MEA.

Solvent	Solvent wt%	Solvent flowrate (kgmol/h)	CO ₂ lean loading (mole _{CO2} /mole _{amin})	CO ₂ rich loading (mole _{CO2} /mole _{amin})	Regeneration Energy (MJ/kgco ₂)	Makeup (kg/h)	Total Power (KW)
MEA	30	73.06	0.2805	0.4995	4.003	20.9211 (water) 0.055 (solvent)	152.62
MDEA/PZ 40 wt%	35 / 5	109	0.2022	0.3601	2.69	-	115
MDEA/PZ 50 wt%	43 / 7	81.27	0.1877	0.3343	2.53	-	107

4.8 Environmental impact assessment of the overall process

The environmental impact assessment results for the overall process estimated in the LCA study are illustrated in **Table 31** and **Figure 28**. The midpoint methodology (in LCA) is an analytical approach that involves assessing the environmental impact of a product or process at an intermediate stage (Huijbregts et al., 2017). They focus on a set of specific midpoint impact categories resulted from using fossil fuels, the chemical utilized during the process, and the ultimate emission to air, water, and land. The impact category potential of the three scenarios of this study using the midpoint methodology is presented in **Table 31**, showing reduction in GWP once PCC is retrofitted to power plant and the findings are similar to reported results (Clarens et al., 2016; Wang et al., 2019; Yin et al., 2017).

The results obtained from different categories can be grouped based on their sources of influence. The first group includes those directly influenced by stack emissions generated during the process. The second group consists of waste effluents, often discharged with wastewater, while the third group is related to emissions caused by the use of chemicals such as solvents and inhibitors.

The influence of the first category, emissions from the process, measured by GWP, OFTEP, and OFHHP illustrated in **Table 31** that shows a tremendous reduction in GWP once PCC is utilized and retrofitted to power plant where the GWP is reduced by 75% once blended solvent (MDEA/PZ) is used. The GWP qualifies the increase of integrated infrared radiative of the greenhouse gases (IPCC, 2013). However, the other indicators, OFTEP and OFHHP show a slight increase once PCC is retrofitted to power plant

compared to the power plant without PCC. The reduction of OFTEP and OFHHP once power plant is driven without PCC is compared to the one with PCC. It found that the effect of chemicals such as solvents during PCC process required more attention to ensure no degradation or loss of solvent as part of the emissions.

The water-related indicators of midpoint methodology include WCP, FWEP and MEP for the three scenarios are illustrated in **Table 31**. The consumption of water increases by 95% once power plant retrofitted with PCC compared to the power plant without PCC. The raise of water demand once PCC process is utilized could be attributed to cooling and heating systems that needed for solvent regenerating. Furthermore, water is needed to prepare aqueous solution of solvent preparation under specific concentration that controls using clean water (Lacy et al., 2015; Matin & Flanagan, 2022). The fresh water ecotoxicity potential measures the impact into freshwater resources due to release of waste such as wastewater from the system (van Zelm et al., 2009). The results of this study (**Table 31**) show that FWEP increases once PCC is utilized and reaches the highest once MEA is used for CO₂ capture. The MEP is similar to FWEP that measures the impact on marine ecosystems during the process life cycle (Helmes et al., 2012). Furthermore, the usage of solvent and other chemicals could increase the risk of MEP specifically once PCC is utilized as shown in **Table 31**.

Indeed, the findings of this study highlighted the need for sustainable water management system and awareness of water conservation through exploring water reuse system that work to reduce the damage of WCP, FWEP, and MEP once PCC unit is utilized.

The third group of midpoint methodology indicators measures the influence due to the utilization of chemicals such as solvent and inhibitors by measuring the damage on TEP. The release of harmful substances from the system boundary is measured by TEP, that mainly focuses on the impact of land and related ecosystem (van Zelm et al., 2009). The results illustrated in **Table 31** of this study show that around 89% increases in TEP once PCC is utilized due to the consumption of solvent and the degradation or side reaction of them. The study of TEP and the effect of PCC raise the need to study further the area of the PCC process for improvement by reducing its environmental impact on land ecosystem through identifying the optimum solvent. However, the result of the study shows that the TEP is reduced by 14% once MDEA/PZ is used for PCC unit compared to the unit using MEA. Hence, further study of the PCC system with MDEA/PZ helps to further reduce the impact of TEP.

Table 31 Environmental impact assessment results for the power plant with and without CC (PCC) using midpoint methodology.

Impact category	Unit	Acronym	Power plant without PCC	Power plant with PCC – using MEA	Power plant with PCC – using MDEA/PZ
GWP	KgCO ₂ eq/yr	GWP	0.906	0.39	0.23
WCP	m ³	WCP	2.40E-03	9.65E-03	5.14E-02
OFTEP	kg NO _x eq	OFTEP	4.90E-04	3.22E-03	2.81E-03
FEP	kg 1,4-DCB	FWEP	1.60E-02	3.46E-02	2.82E-02
MEP	kg 1,4-DCB	MEP	2.30E-02	3.93E-02	3.91E-02
OFHHP	Kg NO _x eq	OFHHP	1.90E-04	3.04E-03	2.79E-03
TEP	kg 1,4-DCB	TEP	0.36	3.37	2.91

The results of the environmental impact assessment through the endpoint methodology identified the comprehensive evaluation of the overall process following the three scenarios illustrated in **Figure 28**. The endpoint indicators include human health, ecosystem quality, and resources depletion are identified for this study's selected scenarios. The result shows that power plant drive without PCC unit has the highest impact on the human health compared to the power plant with PCC (**Figure 28**). It is attributed to the emission of harmful substances that has impact on human health, which reduced once PCC process retrofitted to the power plant. However, **Figure 28** shows that ecosystem quality reached the minimum for power plant drive without PCC compared to power plant drive with PCC. Indeed, ecosystem quality provides insight into the environmental impact of product or process to the biodiversity, habitat disruption, and overall ecosystem health (Roy et al., 2014; Van Zelm et al., 2016). Using solvent with a possibility of degradation or leakage under certain conditions increases the crucial impact on the ecosystem quality. Hence, implementation strategy to minimize the consumption of solvent and control the utilization is required to minimize the adverse effects. Resource depletion, the third indicator of the endpoint methodology measures the impact on the main resources including minerals, fossil fuels, water, and land (Vieira et al., 2016). The resource depletion part in **Figure 28** shows no variation between power plant drive without PCC or with PCC that reached the maximum for the three selected scenarios of this study. The findings support the opportunity to identify other sustainable resources such as renewable energy and hydrogen while considering fossil fuel as a temporary option.

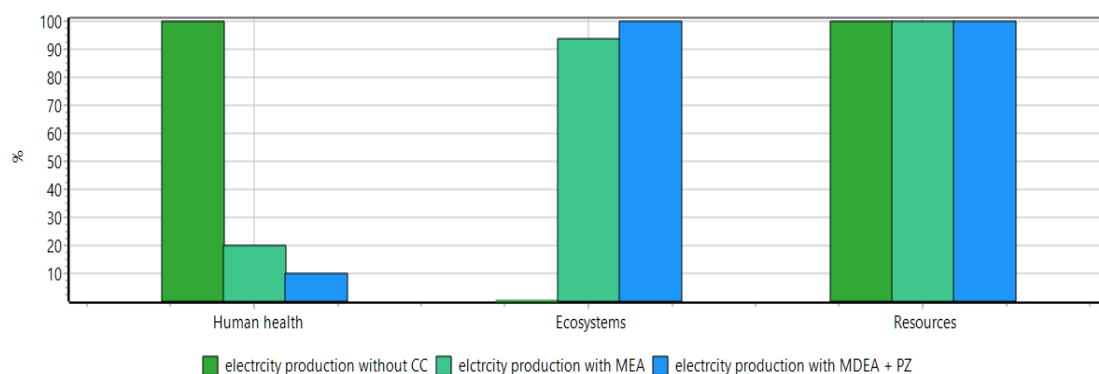


Figure 28 Environmental impact results using endpoint methodology to the overall process.

4.9 Blended MDEA/PZ as a potential solvent option

The aim of this section is to investigate the environmental impact of PCC retrofitting to power plants compared to the power plant without PCC. The two solvents were selected for the PCC unit with the intention to identify the environmental impact of MDEA/PZ compared to MEA. The environmental impact assessments results following the midpoint and endpoint methodologies of LCA show that for some indicators power plant without PCC have a better environmental profile compared to power plant with PCC, while for other indicators the environmental profile shows improvement once PCC retrofitted to power plant (Table 31, Figure 28). However, the environmental profile of the overall process once blended solvent MDEA/PZ is utilized was better than MEA as shown in Figure 29. The finding is attributed to the lower energy demand required for solvent regeneration and the limited degradation identified compared to MEA system. The WCP is the only indicator that is higher for MDEA/PZ compared to MEA by approximately 81%. The literature shows that water demand of MEA is higher than MDEA/PZ (Zhang et al., 2017). The high-water demand of this study could be attributed to the high flow rate of blended solvent using HYSYS (for further details referring to Chapter 3).

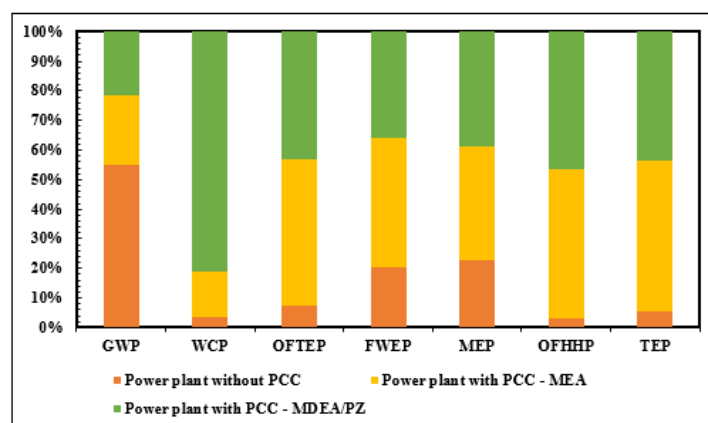


Figure 29 Damage percentage of the impact category using midpoint methodology to the overall process.

The normalization results of this study scenarios following the endpoint methodology, based on normalization factor for Australia system, are illustrated in **Figure 30**. It identified which of the environmental aspects have the most significant contributions to the overall impact compared to a reference value. The result illustrated in **Figure 30** shows that potential impact of human health issues need further consideration than the ecosystem quality and resources depletion. Furthermore, the normalization of resources depletion is consistent with the endpoint methodology findings irrespective of using PCC or otherwise. The resources depletion and ecosystem quality will be highly effected in either case. The endpoint methodology results show improvement for PCC with MDEA/PZ compared to the power plant with or without PCC using MEA.

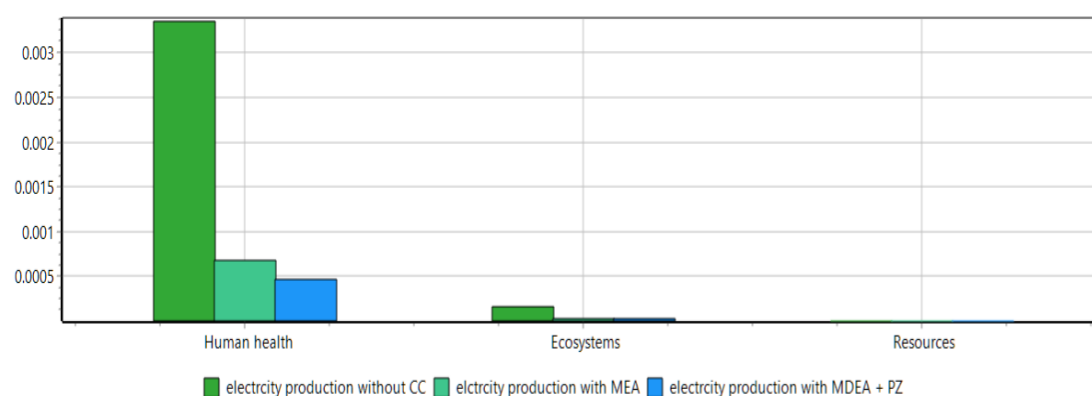


Figure 30 Normalization of environmental impact assessment results.

The GWP over 500-year time horizon (GWP500) is shown in **Figure 31** that measured the carbon footprint follows the IPCC methodology and is expressed in terms of $\text{CO}_{2\text{eq}}$ where all the GHGs are compared based on

their warming potential (Asante-Okyere et al., 2016; Müller et al., 2020). Biogenic carbon refers to the carbon that naturally cycles through various climates, including the atmosphere and ecosystems, and contributes to the carbon footprint of a property (IPCC, 2013). The **Figure 31** shows the reduction of biogenic CO₂ once PCC is retrofitted to the power plant where reduction reached the minimum by 10% once MDEA/PZ is utilized. Hence, the PCC system with MDEA/PZ shows a better performance in terms of carbon footprint compared to PCC system with MEA.

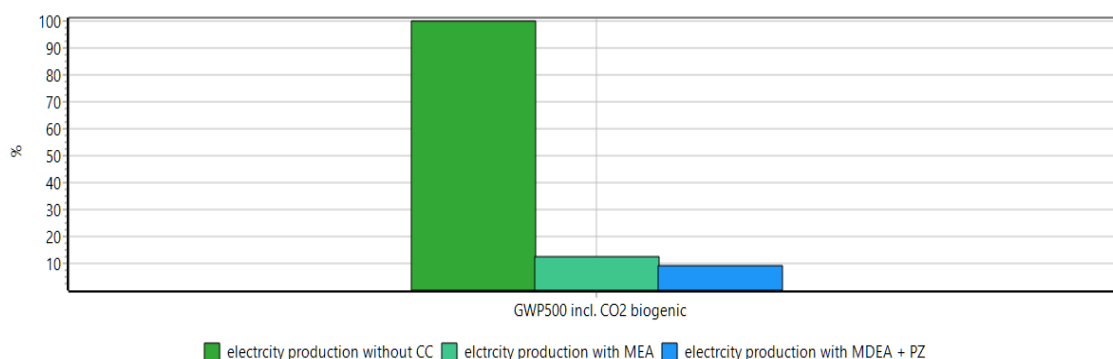


Figure 31 Impact assessment result using IPCC methodology to the overall process.

The findings from related studies in the literature are illustrated in **Table 32**. The findings are categorized based on the scope and boundary of each system, the sources of data, and the findings of each study. Indeed, the reviewed works of LCA for power plant utilized PCC using MEA was identified, however, the LCA of PCC with MDEA/PZ is not identified. Furthermore, the findings of the related study came up with similar conclusion to this study that using PCC reduces the GWP and that insists on considering PCC as a key solution to reduce CO₂ emissions. The reduction of GWP in this study is similar to literature studies that is illustrated in **Table 32**. Although the reviewed articles show an increase in the other environmental indicators once PCC is utilized, that increase is lower once MDEA/PZ is used for PCC process. Therefore, the reduction in GWP occurs despite the implementation of CC techniques, and environmental damages become apparent once these techniques are put into practice. However, the damage to environment was not same for all the CC techniques. This study demonstrates that selecting the optimal solvent has a significant impact on either reducing or increasing environmental damage. Based on the findings, MDEA/PZ is considered the optimal option.

Table 32 Comparison between this study and related studies in literature.

No.	Scope and system boundary	Data quality and sources	Findings	Reference
1	Cradle to gate LCA for power plant with and without PCC using MEA by following midpoint methodology	Data provided from Tae'an coal-fired power plant, literature, and Ecoinvent database.	- Using PCC retrofitted to power plant is an effective solution to capture CO₂ - GWP reduced by 78%, while the other environmental indicators are increased once PCC utilized	(Jung et al., 2022)
2	Cradle to grave for three different coal power plants system with and without PCC using MEA	Data of the power plant are assumed to be for a pulverized coal fired operates in the Netherlands, while the rest taken from Ecoinvent database.	- Greenhouse gas emission reduced by 71 to 78% - Increases the categories of human toxicity, ozone layer depletion and fresh water ecotoxicity once PCC is utilized	(Koornneef et al., 2008)
3	Cradle to grave of power plant includes CCSU along with alternative energy as an option using PCC with MEA	The study taken as a reference case in Canada, the data obtained from Ecoinvent database and published literature following ReCiPe midpoint method.	- Global warming potential was the lowest indicators once CCSU system implemented - Human health risk indicators drop slightly once CCSU utilized, however, resource depletion indicator was high even with CCSU utilization	(Kotagodahetti et al., 2021)
4	Cradle to crave life cycle	The study was for a model of	Power plants Retrofitted with	(Odeh & Cockerill, 2008)

	assessment of power plant drives by different fuel without and with PCC using MEA	power plant in UK while the data are obtained from literature and Ecoinvent database.	PCC enable reduction of CO ₂ emissions. • GWP reduced by 75 - 84%, while the other environmental indicators are increased once PCC utilized	
5	Cradle to gate life cycle assessment of power plant without PCC compared to power plant with PCC using MEA as one option and MDEA/PZ as another option	The data for the LCA is taken from the simulated HYSYS model where the data related to coal process are taken from Ecoinvent v3.5 database.	• PCC retrofitted to power plant promises solution to capture CO₂ • GWP reduced by 75%, while the other environmental indicators are increased once PCC utilized	This work

4.10 Conclusion

This thesis studies the environmental impact of PCC process retrofitted to a 350 MW coal-fired power plant. The study involves investigating the highly significant variables towards reducing the solvent regeneration energy demand, simulation and modelling of PCC plant to investigate the synergistic effect of blended solvent of MDEA/PZ. While the environmental impact of the process specification under the lowest energy demand was investigated in the last part.

Stepwise multi-regression analysis was conducted on a pilot plant solvent-based PCC process to develop a linear model, assessing the impact of seven key variables on energy demand for solvent regeneration and CO₂-rich loading. Only three variables out of seven namely solvent concentration, inlet CO₂ concentration, and lean CO₂ loading have strong significant effect on the regeneration energy. Three show a strong significant effect toward the capture efficiency including the CO₂ desorption rate, lean CO₂ loading, and lean temperature.

The models were validated against multi-regression assumptions. The calculated VIF as part of linearity assumption for the regeneration energy were 1.496, 1.772, and 1.315, while the calculated VIF for the CO₂ rich

loading were 1.212, 1.035, and 1.213. The VIF values less than 10 are accepted and validate the multi regression assumption. The validation test proves the ability of the obtained models to be used for predicting various scenarios.

The linearity model for solvent regeneration energy and CO₂ rich loading was found able to be used for predicting solvent regeneration energy and CO₂ rich loading values for specific scenarios. The linearity model's predictive ability was tested using literature values for solvent regeneration energy and CO₂-rich loading.

The linearity model showed a 0.78% error for predicted CO₂-rich loading and a 2.47% error for solvent regeneration energy compared to literature values. These results confirm the model's suitability for predicting optimal values, reducing effort and cost in process design.

A steady-state rate-based model for the MEA solvent-based PCC process was developed using HYSYS and validated against pilot-scale data from CO₂ capture at the 350 MW Tarong coal-fired power station in Queensland, Australia. The model predictions matched experimental data within $\pm 8\%$. Key variables for validation included CO₂-rich loading, solvent regeneration energy, and CO₂ absorption rate. HYSYS was proved to be suitable for PCC modeling, with one experiment, consuming 4.003 MJ/kg_{CO2} for solvent regeneration, selected as the reference for simulation. The simulation aimed to explore the synergistic effect of MDEA/PZ blend in reducing solvent regeneration energy in the PCC process. Results showed that the combined concentration of 43 wt% MDEA /7 wt% PZ (50 wt% total) required the lowest regeneration energy of 2.53 MJ/kg_{CO2}, that is less by 32.5% compared to the 40 wt% mixture case. This energy savings was achieved without altering process configurations. Minimizing PZ concentration, as noted in literature, reduces solvent degradation and the need for solvent makeup, mitigating potential environmental impacts.

Viscosity drops by 40% at 50 wt% combined solvent concentration, and this attributed to the synergistic effect and an increase in temperature within the system. Following the midpoint methodology of the LCA to investigate the emissions to air, water and land, shows the GWP is reduced by 75% once PCC is utilized. This is attributed to the reduction of CO₂ emissions due to the capture process. However, the other indicators such as OFTEP and OFHHP show a slight increase once PCC is retrofitted to power plant, being attributed to the effect of usage of solvents. More attention is required to ensure no degradation during the process or loss of solvent and scape with the flue gas that is free of CO₂.

Water consumption increases by 95% when a power plant is retrofitted with PCC, due to its need in cooling, heating, and solvent preparation. Implementing sustainable water management and exploring water reuse systems is crucial to mitigate the impact on WCP, FWEP, and MEP when using PCC

The technical assessment of PCC under standard configuration shows MDEA/PZ exhibits high efficiency and less energy demand for solvent regeneration compared to MEA. The LCA study showed that MDEA/PZ has a better performance and environmental profile than MEA, with regard to lower energy demand, solvent consumption, and negligible solvent degradation with 43 wt% MDEA and 7 wt% PZ.

The synergistic effect of MDEA and PZ has proven to be an effective option for enhancing CC efficiency.

4.11 Recommendation for future research

The following are recommended for the performance and environmental evaluation of the solvent-based PCC process.

- It is recommended to assess the economic feasibility of solvent-based PCC process using MDEA/PZ under different process conditions. The capital and operational costs need to be evaluated and find out the possible revenue and financial incentives to reduce the cost and increase the profit.
- The technical assessment of the PCC process shows that the highest energy consumption occurs during solvent regeneration in the stripper column, which negatively affects power plant efficiency. Using the MDEA/PZ solvent combination reduces energy demand by 36.8% compared to MEA, with further potential improvements through optimized process configurations or renewable energy for regeneration. Solar energy is being explored as a green alternative. Additionally, optimizing the absorber and stripper designs could improve efficiency.
- Investigating the scaling up of the PCC model to a larger plant is essential to assess the impact of these design changes on energy consumption and efficiency.
- This thesis explores the synergistic effect of the solvents MDEA and PZ at various concentrations to determine the optimal levels that minimize energy demand for solvent regeneration while securing process efficiency. It recommends investigating the addition of a third solvent to the MDEA/PZ blend, aiming to achieve the same efficiency with lower energy consumption and environmental impact.
- This study assumes a linear model for the PCC processes using MDEA/PZ and MEA under steady-state conditions. However, variations in electricity demand due to changes in power generation inputs and outputs can lead to discrepancies and interactions among process variables, posing challenges to dynamic process behavior. Therefore, further investigation into the effects of process variation on energy demand, efficiency, absorber and stripper design is recommended.
- It is recommended to design an appropriate water management system for the PCC process. This design should investigate the quality of water used, the quantity and quality of water discharge, and physical and

chemical treatment methods for wastewater to maintain quality and prevent environmental damage.

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Appendices

Appendix 1 Data of the dependent and independent variables for PCC pilot test.

x_1	x_2	x_3	x_4	x_5	x_6	x_7	y_1	y_2
lean temperature	lean MEA Concentration	lean CO ₂ loading	inlet flue gas temperature	inlet gas pressure	inlet CO ₂ volume%	CO ₂ desorption rate	Rich CO ₂ loading	regeneration energy
31.7	25.1	0.279	51.7	106.1	11.9	71.6	0.466	4.48
31.4	24	0.294	50.5	105.6	11.8	71	0.48	4.45
31.3	27.9	0.284	48.1	106.3	11.8	70.6	0.469	4.35
35.5	25.5	0.28	51.3	106.6	12.1	73.6	0.471	4.5
33.9	31.6	0.314	53.6	107.5	13.4	72.6	0.481	4.33
34.8	29.6	0.333	54.2	107.3	13.5	76.8	0.486	4.51
35.3	29.2	0.347	55.8	107.3	13.5	76.7	0.488	4.6
33.5	30.3	0.316	65.5	107.3	12.8	76	0.494	4.49
32.4	33.5	0.254	66.1	108.7	12.9	93.9	0.494	4.01
37.5	34	0.278	63.9	108.3	13.2	94.2	0.489	4.04
35.6	29.3	0.414	49.8	107.2	12.7	94.3	0.525	4.55
38.7	32.3	0.285	48.3	106.7	12.2	73.3	0.486	4.01
38.7	32.8	0.291	48.8	106.6	12.2	71.1	0.5	4.02
39.4	29.2	0.285	58.3	108.9	11.1	84.3	0.488	4.21
39.3	29.2	0.285	59.4	108.7	11.2	83.6	0.488	4.23

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39.2	28.9	0.28	56.7	108.8	11.2	84	0.491	4.23
39.7	26.2	0.314	61.8	108.3	11.1	82.4	0.499	4.46
40.5	27.4	0.288	60.7	108.5	11	84.2	0.503	4.29
39.6	28.1	0.295	60	109	11.6	84.6	0.511	4.23
39.4	28.3	0.297	56.1	108.8	11.6	84.1	0.512	4.24
39.6	28.5	0.283	58.2	108.9	11.6	84.7	0.492	4.19
39.5	28.5	0.283	57.9	109	11.1	84.2	0.492	4.22

Appendix 2 Blended solvent (MDEA/PZ) properties for 40 wt% total.

Case	Solvent wt% (MDEA/PZ)	Solvent flow rate (kgmole/hr)	CO2 lean loading	Viscosity solvent (cP)	Viscosity rich solvent (cP)	Viscosity lean solvent (cP)	CO2 rich Loading	CO2 Absorption rate Kg/h	Makeup (kg/hr)	
									Water	Solvent
1	38 - 2	171.8	0.1326	5.746	4.688	0.948	0.2362	74.793	0	0
2	35 - 5	109	0.2022	6.876	4.846	1.128	0.3601	75.1668	0	0
3	30 - 10	84.5	0.2459	7.512	5.310	1.102	0.4382	74.964	0.2864	0
4	28 - 12	80.75	0.2569	7.218	5.094	1.055	0.4572	74.7864	0.2756	0
5	25 - 15	75.08	0.2689	7.109	4.778	1.024	0.4788	74.9042	0.5513	0
6	20 - 20	68.5	0.2865	6.888	4.080	1.045	0.5099	74.988	3.5112	0.086(PZ)

Appendix 3 Power required for blended solvent (MDEA/PZ) for 40 wt% total.

Case	Solvent wt% (MDEA/PZ)	Power - Pump (kW)	Power - Reboiler (kW)	Power - Compressor (kW)	Power - Cooler (kW)	Total power (KW)	Reboiler Temperature C	Regeneration Energy (MJ/KgCO ₂)
1	38 - 2	0.1211	80.35	15.29	67.28	163.0411	103.4	3.86
2	35 - 5	7.679×10^{-2}	56.24	15.02	43.82	115.157	102.7	2.69
3	30 - 10	5.989×10^{-2}	76.850	14.740	64.280	155.930	106.4	3.691
4	28 - 12	5.701×10^{-2}	84.08	15.18	71.25	170.567	107.7	4.04
5	25 - 15	5.307×10^{-2}	88.02	16.53	73.42	178.023	109.2	4.23
6	20 - 20	4.814×10^{-2}	92.29	19.09	72.77	184.198	111.1	4.43

Appendix 4 Blended solvent (MDEA/PZ) properties for 50 wt% total.

Case	Solvent wt% (MDEA/PZ)	Solvent flow rate (kgmole/hr)	CO2 lean loading	Viscosity solvent (cP)	Viscosity rich solvent	Viscosity lean solvent	CO2 rich Loading	CO2 Absorption rate Kg/h	Makeup (kg/hr)	
									Water	Solvent
1	43 - 7	81.27	0.1877	14.45	8.665	1.605	0.3343	74.6711	0	0
2	40 - 10	73.56	0.2034	15.000	8.890	1.365	0.3621	75.017	0	0.0119(MDEA)
3	35 - 15	65.8	0.2207	14.56	8.019	1.109	0.3927	75.0581	0.4504	0
4	30 - 20	60.18	0.2345	14.39	7.05	1.023	0.4173	74.7632	2.9959	0.0086(PZ)

Appendix 5 Power required for blended solvent (MDEA/PZ) for 50 wt% total.

Case	Solvent wt% (MDEA/PZ)	Power - Pump (kW)	Power - Reboiler (kW)	Power - Compressor (kW)	Power - Cooler (kW)	Total power (KW)	Reboiler Temperature C	Regeneration Energy (MJ/KgCO ₂)
1	43 - 7	6.523×10^{-2}	52.49	13.97	41.46	107.985	102.2	2.53
2	40 - 10	5.922×10^{-2}	74.29	13.79	62.91	151.049	106	3.56
3	35 - 15	5.311×10^{-2}	90.33	15.95	76.04	182.373	109.4	4.33
4	30 - 20	4.842×10^{-2}	94.5	19.48	75	189.028	111	4.55

Appendix 6 Life cycle impact assessment result for midpoint (MDEA/PZ) for 50 wt% total.

Impact	Unit		Midpoint			Midpoint (Normalization)		
		Impact	Power plant without PCC	Power plant with PCC (30 wt% MEA)	Power plant with PCC (47 wt% MDEA/ 7 wt% PZ)	Power plant without PCC	Power plant with PCC (30 wt% MEA)	Power plant with PCC (47 wt% MDEA/ 7 wt% PZ)
Global Warming Potential	yr/KgCO ₂ eq	GWP	0.906	0.39	0.35	1.13E-04	4.86E-05	4.36E-05
Water Consumption Potential	m ³	WCP	2.40E-03	9.65E-03	5.14E-02	9.01E-06	3.62E-06	0.000193
ozone formation terrestrial ecosystem potential	bg NO _x eq	OFTEP	4.90E-04	3.22E-03	2.81E-03	2.76E-05	0.000181	0.000158
Fresh Water Ecotoxicity potential	kg 1,4-DCB	FWEP	1.60E-02	3.46E-02	2.82E-02	6.35E-04	0.00137	0.00112
marine ecotoxicity potential	kg 1,4-DCB	MEP	2.30E-02	3.93E-02	3.91E-02	5.29E-04	0.000905	0.000899
ozone formation human health potential	Kg NO _x eq	OFHHP	1.90E-04	3.04E-03	2.79E-03	9.26E-06	0.000148	0.000136
terrestrial ecotoxicity potential	kg 1,4-DCB	TEP	0.36	3.37	2.91	2.37E-05	0.000222	1.92E-04

Appendix 7 Life cycle impact assessment result for endpoint (MDEA/PZ) for 50 wt% total.

	Endpoint (Normalization)			Endpoint (weighting)		
Category	Power plant without PCC	Power plant with PCC (30 wt% MEA)	Power plant with PCC (47 wt% MDEA/ 7 wt% PZ)	Power plant without PCC	Power plant with PCC (30 wt% MEA)	Power plant with PCC (47 wt% MDEA/ 7 wt% PZ)
Human health	0.00333	0.000658	0.000445	1.33	0.263	0.178
Ecosystems	0.000163	2.12E-05	2.22E-05	0.0654	0.00847	0.00888
Resources	3.14E-06	3.14E-06	3.14E-06	6.27E-04	6.27E-04	6.27E-04

Appendix 8 Life cycle impact assessment inventory for (MDEA/PZ) for 50 wt% total.

Gas	Emissions amount - without PCC (kg/h)	Emissions amount - with PCC (MEA) (kg/h)	Emissions amount - with PCC (47 wt% MDEA/ 7 wt% PZ) (kg/h)
CO ₂	84.8075	9.8089	9.7855
H ₂ O	11.1796	31.4896	10.0662
O ₂	52.2038	52.2001	52.1988
N ₂	340.4091	340.3961	340.3925

الملخص

إن انبعاثات ثاني أكسيد الكربون الناتجة عن حرق الوقود الأحفوري في محطات الطاقة تشكل مساهماً رئيسياً في تغير المناخ. ويشكل احتجاز الكربون وتخزينه حلاً يعول عليه للحد من هذه الانبعاثات في حين يتم تطوير مصادر الطاقة المتجددة الأخرى. ومن بين تقنيات احتجاز الكربون وتخزينه، يعد احتجاز الكربون بعد الاحتراق طريقة واعدة للحد من انبعاثات ثاني أكسيد الكربون من محطات الطاقة القائمة. ومع ذلك، فإن احتجاز الكربون بعد الاحتراق له عيوب، بما في ذلك تكاليف البناء المرتفعة، والطلب المرتفع على الطاقة لإعادة توليد المذيبات، والتأثيرات البيئية المحتملة.

تم استخدام التحليل الإحصائي لتحديد المتغيرات المهمة في عملية احتجاز الكربون بعد الحرق لتقليل طاقة تجديد المذيبات وتعزيز الكفاءة. تم اختيار سبعة متغيرات مستقلة، بما في ذلك درجة حرارة المذيب الهزيل، وتركيز المذيب، وثاني أكسيد الكربون الهزيل، وظروف غازات الاحتراق. كشف التحليل أن تركيز المذيب، وتركيز ثاني أكسيد الكربون الداخل، وثاني أكسيد الكربون الهزيل يؤثر على طاقة تجديد المذيبات. تم التحقق من صحة معادلات الاحصائية ووجد أنها موثوقة، مع أخطاء تنبؤ بنسبة 0.78%. للتحميل الغني بثاني أكسيد الكربون و 2.47% لطاقة تجديد المذيبات.

تم تطوير نموذج قائم على معدل حالة الدراسة لنظام احتجاز الكربون بعد الحرق باستخدام MEA كمذيب وتم التحقق من صحته في Aspen HYSYS باستخدام بيانات المصنع التجريبي. اتفقت تنبؤات النموذج باستخدام HYSYS مع بيانات المصنع التجريبي لعملية احتجاز الكربون بعد الحرق حيث لم يتجاوز الانحراف بين النموذج والنتائج التجريبية $\pm 8\%$. تمت تصميم محاكاة جديدة لعملية الاحتجاز باستخدام المذيب المركب MDEA / PZ تحت تركيز إجمالي مختلف (40% بالوزن و 50% بالوزن) وتركيز فردي مختلف لـ MDEA و PZ. كان الهدف من عملية محاكاة الحجز باستخدام المذيبات المركبة هو التحقيق في التأثير التآزري للمذيبات المختارة مقارنة بـ MEA عند 30% بالوزن. تظهر المحاكاة أن 43% من وزن MDEA و 7% من وزن PZ هي التركيبة المثلى التي تتطلب الحد الأدنى من الطاقة لتجديد المذيب بمقدار 2.53 MJ/kgCO₂ لتحقيق كفاءة عملية بنسبة 88.5% مقارنة بـ 4.003 MJ/kgco₂ لـ 30% من وزن MEA دون الحاجة إلى أي تعديلات على العملية.

تم استخدام تقييم دورة الحياة من المهد إلى البوابة للتحقيق وتقييم التأثير البيئي لعملية حجز الكربون بمجرد إعادة تركيبه في محطة الطاقة باستخدام SimaPro. تم استخدام ثلاث طرق لتقييم دورة الحياة في هذه

الدراسة بما في ذلك نقطة منتصف ReCiPe 2016 (H) ونقطة نهاية ReCiPe 2016 (H) و IPCC 2013 GWP 500. تظهر النتائج من دراسة تحليل دورة الحياة أن محطة الطاقة المجهزة بحجز الكربون بعد الحرق أظهرت انخفاضاً هائلاً في إمكانية الاحتباس الحراري العالمي مقارنة بمحطة الطاقة بدون معدات حجز الكربون. ومع ذلك، وجدت الدراسة أن محطات الطاقة مع أو بدون معدات حجز الكربون تسبب استنزاف الموارد بسبب استخدام الوقود الأحفوري. علاوة على ذلك، تؤثر العديد من الانبعاثات البيئية على موارد المياه على وجه التحديد وتصل إلى الحد الأقصى بمجرد استخدام MEA لحجز الكربون مقارنة بـ MDEA / PZ المطلوب هو تصميم نظام إدارة المياه لإدارة استخدام المياه أثناء عملية حجز الكربون. ومن ثم، تظهر الدراسة التأثير التآزري لـ MDEA و PZ لالتقاط ثاني أكسيد الكربون باستخدام عملية حجز الكربون الموصى بها، مما يظهر القدرة على حل عيوب عملية حجز الكربون المتعارف عليها من الناحية الفنية والبيئية ويمكن اختيارها كواحد من حلول التخفيف للحد من انبعاثات ثاني أكسيد الكربون. إلا أن الدراسة لم تشتمل على التقييم الاقتصادي للنظام والتباين في عملية محطة توليد الكهرباء وكذلك مراحل النقل والتخزين الخاصة باحتجاز الكربون واستخدامه أو عملية تخزينه.

جامعة البحرين



برنامج البيئة والتنمية المستدامة

كلية العلوم

التقييم البيئي لعملية احتجاز الكربون المنبعث بعد حرق الوقود في محطات الطاقة: تقييم دورة الحياة

أطروحة مقدمة كجزء من متطلبات الحصول على درجة دكتوراة الفلسفة في البيئة والتنمية المستدامة

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