Techno-economic analysis of the production of pure CO_2 from flue gases via the monoethanolamine and ammonia capture processes

by

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Abstract

Carbon dioxide (CO_2) emissions from fuel combustion have contributed to the overall greenhouse gas emissions worldwide. Carbon capture is a promising technique that could reduce the emission from power plants with the ever-increasing energy demand. Research into the utilization of CO_2 from flue gas is in advanced stage and therefore, there is a need to develop the baseline cost for separation and purification of CO_2 from various flue gases. In this study, a detailed process model was developed for carbon capture from flue gases produced in coal-fired power plants and natural gas combined cycle (NGCC) plants using monoethanolamine (MEA)-based solvent. In addition, a detailed techno-economic assessment of the whole process chain of producing pure CO_2 using ammonia and monoethanolamine (MEA) based solvents for the flue gas from coal-fired power plants is evaluated and compared. The main operations characterized in this study are the absorption of carbon dioxide with the MEA solution, the regeneration of the MEA solution in the stripper for its reuse and separation, and the compression of CO_2 to the desired exit pressure range. The costs of producing pure CO_2 through the capture process for a coal-fired power plant with a capacity of 500 MWe at capture rates of 90%, 95%, and 99% are \$60.65, \$61.57, and \$62.16 /tonne $CO_{2,}$ respectively; and for an NGCC plant with a capacity of 555 MWe and the same capture rates are \$79.47, \$84.44, and \$87.07 /tonne $CO_{2,}$ respectively. In comparison, ammonia based solvent, the cost of producing pure CO_2 at 90% capture rate for coal flue gases with plant capacity of 500 MWe is 48.42 \$/tonne CO_2, respectively. Furthermore, the regeneration energy obtained for a 90% capture rate using ammonia solvent is 2.46 GJ/tonne CO_2 while that of MEA solvent is 4.62 GJ/tonne CO_2. Though there is a significant progress in the use of chilled ammonia process in terms of successful pilot testing, the technical challenge such as ammonia slip should be addressed in order to gain economic acceptability. The comparative results show that ammonia-based solvent is more cost effective than the amine-based solvent due to the high regeneration energy for the latter process. The sensitivity analyses show the production cost of pure CO_2 is highly sensitive to the capital cost and internal rate of return (IRR). The results of this study will be of interest to investors in chemical processes that intend to use pure CO_2 as feedstock and also for jurisdictions such as Alberta where carbon tax policy is already implemented.

Preface

This thesis is an original work by Elvis Ehizojie Ibadin supervised by Dr. Amit Kumar.

Chapter 2 of this thesis was submitted to the journal *International Journal of Greenhouse Gas Control* as “Techno-economic assessment of the production of pure CO_2 from flue gases via the monoethanolamine (MEA) capture process” by Elvis Ibadin, Adetoyese Olajire Oyedun and Amit Kumar.

Chapter 3 of this thesis will form part of the article to be submitted to the journal of CO_2 Utilization as “Techno-economic assessment of the production of pure CO_2 from flue gases via the ammonia capture process” by Elvis Ibadin, Adetoyese Olajire Oyedun and Amit Kumar.

In these chapters, I was responsible for data gathering, model development and interpretation, and the composition of manuscript. Adetoyese Olajire Oyedun did an extensive review of the developed model, assessed the results and edited the manuscript. Amit Kumar was the supervisor of this work and contributed immensely in research direction, formulation of the concept, analyzed the obtained results and edited the manuscript.

Dedication

I would like to dedicate this work to my lord and savior Jesus Christ. Also, to the blessed memory of my brother, Mike Ibadin.

Acknowledgements

First, I would like to express my sincere gratitude to my supervisor, Dr. Amit Kumar, for his guidance, recommendations, and instruction during the course of the research. Dr. Kumar's vast research experience and his passion for quality work, together with his willingness to share knowledge, were an inspiration throughout the study. For me, it was a great privilege to work under his supervision. In addition, I would like to acknowledge Dr. Adetoyese (Toye) Oyedun for his constructive feedback and assistance throughout this work. Dr. Oyedun's encouragement and motivation went a long way toward the accomplishment of this research.

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List of Abbreviations

APEA	Aspen Process Economic Analyzer
CAP	Chilled ammonia process
CCS	Carbon capture and storage
CCU	Carbon capture and use
CO_2	Carbon dioxide
DCC	Direct contact cooler
DCF	Discounted cash flow
e-NRTL	Electrolyte non-random two liquid
EOR	Enhanced oil recovery
FGD	Flue gas desulfurization
G&A	General and administrative costs
GHG	Greenhouse gas
Gj	gigajoule
IEA	International Energy Agency
IRR	Internal rate of return
MEA	Monoethanolamine
MWe	Megawatts electric
NETL	National Energy Technology Laboratory
NG	Natural gas
NGCC	Natural gas combined cycle
NH_3	Ammonia
PCC	Pulverized coal combustion

ppmv	Parts per million by volume
PV	Product value
SCPC	Supercritical pulverized coal
TPEC	Total purchased equipment cost
TPI	Total project investment

Chapter 1: Introduction

1.1 Background

Worldwide energy consumption and associated carbon dioxide (CO_2) emissions are increasing [1]. From 1990 to 2015, CO_2 emissions from fuel combustion saw a 57.5% increase [2]. The steady rise in energy demand is as a result of worldwide economic growth and development. For example, in 2015, fossil fuel accounts for 82% of the world primary energy supply; therefore, fossil fuels plays a key role in the increase of CO_2 emissions [2]. An Intergovernmental Panel on Climate Change (IPCC) report predicted a global rise in temperature to about 5.8^{o}C between 1990 and 2100 with continued emissions from fossil fuel use [3]. The IPCC also concluded that a significant decrease in global GHG emissions is needed in order to maintain the average increase at 2-2.4 ^{o}C above pre-industrial levels [4]. In other words, the year 2000 scenario CO_2 emissions need to be reduced by half by the year 2050. There are several ways to reduce GHG emissions, such as energy conservation and efficiency measures, renewable energy use, fossil fuel switching, and carbon capture and storage (CCS).

CCS is considered one of the technologies that can significantly reduce GHG emissions from fossil fuel use [5, 6]. With the right technological developments, CCS could be a viable means of reducing overall CO_2 emissions [6]. The International Energy Agency (IEA) reported that CCS technology could reduce cumulative emissions by 13% (or 6 billion tonnes of CO_2 emissions captured and stored annually) by the year 2050, thereby limiting the increase in global temperature to 2^{0}C [5, 7]. Though CCS is usually regarded as the solution to reducing GHG emissions [8], it has several technical and economic challenges, such as high capital cost, public acceptance, lack of strong regulations in many countries, increased electricity cost, etc. [9, 10].

Carbon capture and utilization (CCU) is thus considered an attractive alternative as it not only mitigates CO_2 emissions but also uses the captured CO_2 for commercial products instead of permanently storing it [9-11]. The revenue generated from the use of pure CO_2 as raw material feedstock could offset the high cost of capture and sequestration.

The CCU option is considered a technology that has significant applications, such as the reuse of CO_2 in chemical industries. [12]. Though CO_2 use as feedstock has long been a practice, only a few applications have been realized [13]. Nevertheless, with considerable public interest in the effects of global warming, attention is being given to reducing anthropogenic CO_2 emissions [13], and this has led to technological advancements in CCU technologies recently.

There are three main technologies for carbon capture such as oxy-fuel combustion, pre-combustion capture, and post-combustion capture. Oxy-fuel combustion involves the removal of nitrogen from air in an air separation unit [6]. The fossil fuel is combusted in pure oxygen and the recycled flue gas is used to control temperature rather than air. The separation of oxygen from air is energy intensive, and oxy-fuel technology is still in the demonstration stage and requires high purity oxygen [14]. Pre-combustion capture, on the other hand, involves the reaction of flue gas first with oxygen and/or stream to produce “syngas”; the resulting CO_2 and hydrogen are subsequently processed in a shift reactor to produce a mixture of hydrogen through gasification or a reforming process [14]. Post-combustion capture involves CO_2 capture from fossil fuel flue gases from an existing power plant [15]. Post-combustion capture can be retrofitted into an existing coal- or natural gas-fired plant or simply built with a new plant. Of these technologies, post-combustion capture is considered mature. The Boundary Dam in Saskatchewan, Canada is the largest post-combustion plant in Canada and the first plant in the world to use post-combustion technology. It has been in operation since 2014 with a net capacity of 110 MWe [7].

There are already several technologies for the separation and capture of CO_2 from flue gases in power plants; these include chemical absorption, gas separation, distillation, and physical processes [16]. Among the proposed technologies, the amine-based process is in the commercial stage and has been widely used for over a decade in large-scale chemical industries [14, 17, 18]. Amine technology for CO_2 separation has been used since the 1970s and has improved significantly over time [7]. However, there are challenges with the use of mono-ethanolamine (MEA) as a solvent, such as increased thermal energy during capture and CO_2 compression, solvent degradation rates, and corrosion of equipment [16]. Thus, a more energy-friendly, less corrosive solvent, and cost-effective liquid solvent is needed for the carbon capture process; ammonia is favored over amine. A more detailed discussion on use of amine as a solvent is discussed in subsequent chapter.

Several authors have highlighted the merits of using ammonia for CO_2 capture rather than an MEA solvent specifically in terms of the lower energy requirement [19-21]. Ammonia (NH_3) is well known to be able to withstand degradation in the presence of flue gases such as NO_x and SO_x [22]. An ammonia solvent can also operate at high pressure, which helps lower compression costs during the capture process [23]. The use of ammonia solvent comes with drawbacks, however, the main one being the ammonia slip [24-27]. Care must be taken to avoid ammonia volatility, as ammonia is hazardous to the environment and to people. Some ways to control solvent slip are to decrease the ammonia concentration in the solvent, operate at lower temperatures (preferably 0-10 ^{o}C), and have a high CO_2 loading [21, 27].

Earlier studies have assessed the economics of MEA- and NH_3-based CO_2 capture with a particular focus on the CO_2 avoided cost [28-32]. For example, Li et al. [29] conducted a techno-economic analysis of aqueous, ammonia-based post-combustion capture at 650 MWe capacity. Their findings show that the cost of CO_2 avoided with an ammonia-based process is US$ 67 /tonne CO_2 and the cost for an MEA-based capture process is US$ 86.4 /tonne CO_2. An economic analysis performed by Ciferno et al. [20] on CO_2 capture with an ammonia-based solvent gives the overall cost of CO_2 avoided as 35.37 $/tonne CO_2 and 61.57 $/tonne CO_2 for an MEA process.

The avoided cost is the cost of CO_2 emissions to the surroundings while producing equal amounts of product from a reference plant, usually expressed in dollars per tonne of CO_2 not emitted in reference to the source [33]. However, our focus is on the cost of producing pure CO_2 from coal- and natural gas-fired plants. The CO_2 is considered an industrial commodity for re-use purposes, largely as raw material to other industries for use in enhanced oil recovery (EOR), urea fertilizer, beverage carbonation, food processing, pharmaceuticals, etc.

Many techno-economic assessments of CCS have been conducted [29, 30, 34, 35]; however, little focus has been put on carbon capture from natural gas- and coal-fired power plants where CO_2 is treated as a commodity. There are also few comparative techno-economic analyses of CO_2 production costs from MEA- and ammonia-based post-combustion capture from coal-fired plant flue gases. Therefore, a study is needed on the cost of producing pure CO_2 from flue gases using amine solvent, at the same time comparing the economic effects of the use of different solvents such as MEA and ammonia on overall CO_2 production costs.

1.2 Research objectives

The overall purpose of this research is to carry out a detailed techno-economic assessment of the costs of producing pure CO_2 from flue gases using an MEA-based process and to evaluate the economic benefits of using different solvents such as ammonia and MEA in order to determine the costs of CO_2 production. The specific objectives of this study are to:

- Develop a baseline simulation model for producing pure CO_2 with MEA from coal- and natural gas-fired plants.
- Develop techno-economic assessment models to evaluate the costs of producing pure CO_2 with MEA from coal- and natural gas-fired plants.
- Conduct a detailed, comparative study on the costs of producing pure CO_2 from a coal-fired power plant with MEA- and NH_3-based capture processes.
- Evaluate the effects of the use of MEA and NH_3 solvents on the overall costs of producing pure CO_2 from a coal-fired power plant in $/tonne CO_2.
- Conduct sensitivity analysis to study the influence of various technical and economic parameters on the costs of producing pure CO_2 with MEA- and ammonia- based capture processes.
- Perform uncertainty analyses on both processes.

1.3 Scope and limitation of the study

In this study, we developed a base model for pure CO_2 production from flue gases from coal- and natural gas-fired power plants using MEA and compared the economic benefits of using ammonia and MEA solvents to produce pure CO_2 from flue gas from a coal-fired power plant. The developed model was used to carry out the techno-economic assessment of both MEA- and ammonia-based capture. In CO_2 capture, several solvents are used, such as potassium carbonate, diethano amine (DEA), piperazine (PZ), etc.; however, MEA and ammonia were selected in this study to determine the cost of producing pure CO_2. These solvents were chosen because MEA is commercialized and ammonia not only is more economical in terms of CO_2 production cost but also requires less regeneration energy.

Some of the key assumptions made during the course of model development are:

- Flue gas from the coal-fired power plant has undergone flue gas desulfurization (FGD) in a scrubber to remove sulfur dioxide (SO_2).
- Flue gas was free of NO_x.
- The equilibrium stage assumed for both the absorber and the stripping column.
- The Murphree efficiency was assumed to be 1 and 0.1 for the MEA- and NH_3-based processes, respectively, indicating that the liquid and vapor phases on the tray are well mixed.

In the techno-economic model, the location of the hypothetical plant is in Alberta, Canada. Therefore, Alberta-specific rates were used in calculations. The cost of electricity production was not considered in this study.

1.4 Organization of the thesis

This thesis has four chapters and is in paper-based format; each chapter is independent from the others.

Chapter 1 outlines the thesis background, summarizes the overall objectives of the study, and gives the scope and limitation of the study.

In Chapter 2, the simulation model for production of pure CO_2 from flue gases from coal- and natural gas-fired power plants with the use of MEA as a solvent is described. Also a discussion on the development of techno-economic models for these pathways is discussed.

In Chapter 3, the simulation model for production of pure CO_2 from flue gases from coal power plants with the use of ammonia-based solvent is described. Also a discussion on the development of techno-economic models for these pathways is discussed.

Chapter 4 concludes this research and outlines recommendation for future work.

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Chapter 2: Techno-economic assessment of the production of pure CO_2 from flue gases via the monoethanolamine (MEA) capture process[1]

2.1 Introduction

Fossil fuel-based greenhouse gas (GHG) emissions have increased worldwide since the late 19th century [1]. From 1970 to 2011, fossil fuel-based GHG emissions increased by about 90%. Fossil fuel combustion and industrial processes made up 78% of the overall GHG emissions from 1970 to 2011 [1]. The International Energy Agency (IEA) reported in 2008 that about 69% of CO_2 emissions, a major GHG and 60% of all GHGs are energy-related emissions, [2]. In Canada, the total estimated GHG emissions nationwide were 20% higher in 2002 than the 1990s level of 609 Mt, and 81% of the emissions were from the energy sector [3]. Moreover, the US DOE has projected that until 2030, 95% of emissions will be from coal [4]. GHG emissions need to be reduced globally to avoid adverse impact on the environment. One approach to GHG emissions reduction that has been widely discussed in the literature is carbon capture and storage.

Carbon capture and storage (CCS) is considered a promising technology that can drastically reduce GHG emissions from fossil fuel use [5]. CCS is predicted to reduce cumulative GHG emissions by about 19% by 2050, thus maintaining the 2 ^{o}C global temperature increase and avoiding dangerous climate change [5, 6]. In addition, combined with GHG emissions reduction

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incentives, CCS is considered one of the promising routes to mitigate CO_2 emissions [2, 7]. Several technologies exist for the separation of CO_2 from flue gases in power plants; these include chemical absorption, gas separation, distillation, and physical processes [8]. Out of the technologies proposed to capture CO_2 from power plants, amine-based processes are in the commercial stage and have been widely used for over a decade in large-scale chemical industries [9-11]. Although CCS is expected to mitigate global warming, it has technical and economic challenges including its high cost, which has affected its large-scale deployment [12, 13]. The high cost is mainly due to increased energy use during the capture process and CO_2 compression. In addition, legal questions and public acceptance need to be addressed [10]. Because of these concerns, carbon capture and use (CCU) is considered an attractive option not only to reduce CO_2 emissions but also to convert the captured CO_2 into commercial products instead of permanently storing it; CCU also provides a revenue stream that could offset the costs of capture and sequestration [12-14].

Captured CO_2 is traditionally sequestered underground. However, the chances of using the captured CO_2 for enhanced oil recovery (EOR) are being explored [15]. The end products of CO_2 use (i.e., chemicals, materials, and transportation fuels) are more desirable and in the long term provide a better and longer term solution than CCS [12]. In other words, CO_2 is an essential commodity that could be converted to valuable feedstocks for the manufacture of various consumer products [16]. There are techno-economic assessment studies for CCS; however, in these studies little emphasis has been placed on the cost of producing pure CO_2 for further use. While several authors [17-20] have conducted economic analyses of avoided CO_2 costs in coal and natural gas power plants, there are very limited information on the production costs of pure CO_2 through the capture process with the end goal of considering it a feedstock for CO_2 use and reflects the purity of CO_2. Therefore, a baseline model is needed to develop this information. This model should be data-intensive in order to generate meaningful results; and an assessment of the capture costs from various flue gases from power plants is also needed. This type of information could be used by the decision makers in making investment decisions and policy formulation.

Several CCS cost assessments are available in the literature. A number of studies include an economic analysis of the CO_2 avoided in coal and natural gas power plants estimated as abatement costs [21-23]; however, there are few techno-economic assessments for the production of pure CO_2 through the capturing process with the sole aim of using the produced CO_2 as feedstock for subsequent chemical conversion processes. Some of the CCS studies are discussed.

Sipocz and Tobiesen conducted a thermodynamic and economic evaluation of a 440 MWe natural gas combined cycle power plant with an integrated CO_2 removal plant using MEA as the absorbent [21]. They assessed the techno-economics of the basic MEA technology and exhaust gas recirculation (EGR) integrated with natural gas combined cycle (NGCC) and concluded that EGR in combination with a lowered specific reboiler duty helps reduce operational and capital costs. Mathieu and Bolland did a technical and economic comparison of different systems of CO_2 capture in NGCC. Their results show that the cost of CO_2 avoided in natural gas was $125.34[2] /tonne; this figure is almost twice the coal post-combustion capture cost of $51.52 tonne CO_2 [24]. A policy report by the European Academies' Science Advisory Council (EASAC) stated that for CO_2 capture to be economically attractive, the capture costs would have to be around $69.37 /tonne CO_2 for coal-fired power plants and around twice that for gas-fired power plants, and these costs would have to be guaranteed for the long term [25]. The main

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[2] All cost values used in this study were converted to 2016 USD and the base year in this study is 2016.

difference between capturing CO_2 for further reuse purposes and capturing it for sequestration is the role of energy. In the first instance, energy is a commodity; in the second, the use of energy generates CO_2 emissions that are considered avoidable [26]. In CCS, moreover, the emphasis is on reducing inputs rather than on commercial purposes. Several studies have used the two costs (CO_2 avoided cost and CO_2 captured cost) interchangeably, and this has led to unclear differentiation between the two measures [10]. However, this study focuses on CO_2 as a commodity and hence production cost will be used in this study to describe the capture cost. The studies summarized above show that considerable work has been done in the area of techno-economic assessment of CCS and very little on carbon capture from natural gas- and coal-fired power plants that consider CO_2 a commodity, and thus limited focus has been put on the cost of producing CO_2 for later use. This is the major research gap that will be addressed in this study.

The overall objectives of this research are to develop a baseline simulation model for the production of pure CO_2 from coal and natural gas-fired power plants using MEA as an absorbent and to develop a techno-economic assessment model to compare the production costs of various flue gases. The specific objectives are:

- To develop process simulation models for CO_2 separation and purification from flue gases from the coal and natural gas power plants.
- To develop a techno-economic model to determine the cost of producing pure CO_2 (\$/tonne CO_2) from coal-fired power plants.
- To develop a techno-economic model to determine the cost of producing pure CO_2 (\$/tonne CO_2) from natural gas-fired power plants.
- To compare the costs of CO_2 production from the flue gases of coal- and natural gas-fired power plants.

- To study the effects of changing various parameters on the overall cost of CO_2 production and to conduct uncertainty analysis.

2.2 Process description

2.2.1 Model development

The process base model developed in this study uses flue gases from a 500 MWe coal-fired power plant and gas turbine emissions from a 180 MWe NGCC power plant that was scaled up to 555 MWe so that the model results could be compared with published data. The process conditions of the first plant were derived from research carried out by Fisher et al. [27] and data for the second were from a commercial scale power plant in western Canada [28]. The model for CO_2 capture from both flue gases was developed in Aspen Plus V8.8 [29], and the detailed process flow diagram for the simulation is shown in Figure 2.1.

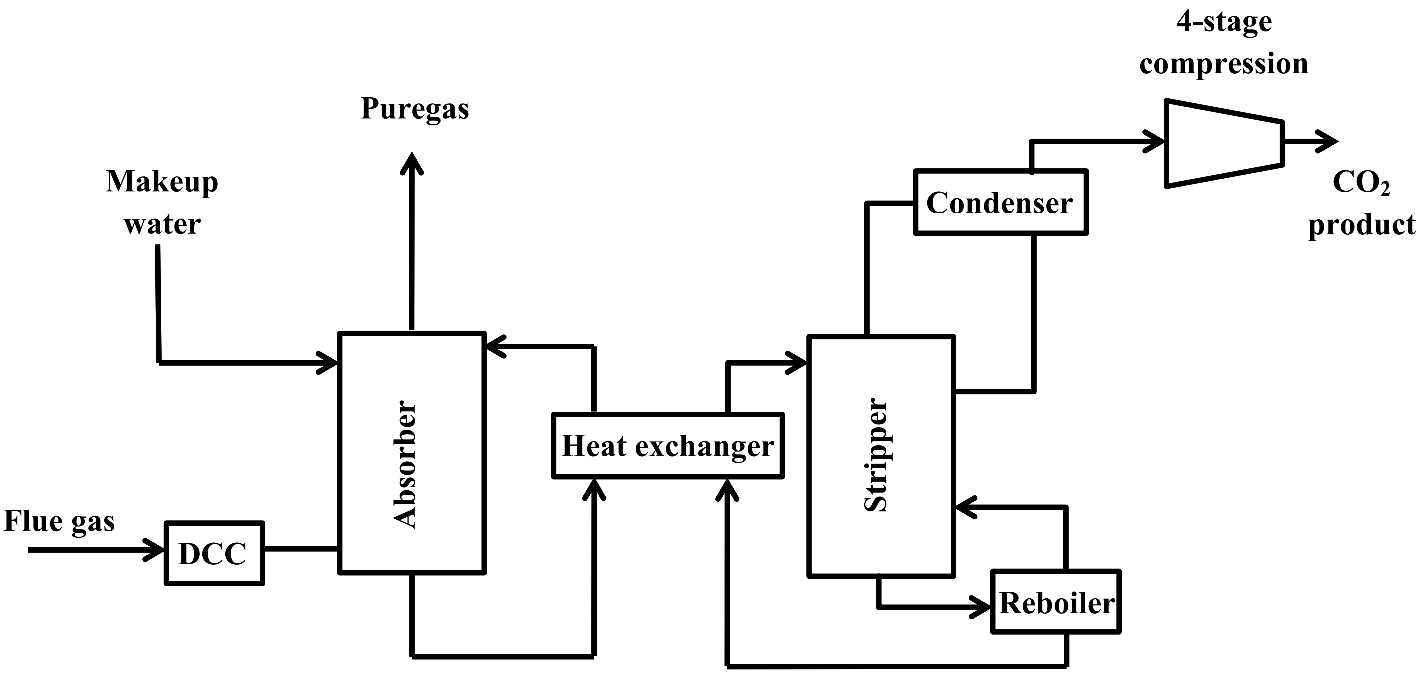

Figure 2.1: Process flow diagram for the developed simulation model

Some key assumptions used in this model are:

- Flue gas from the coal power plant has undergone flue gas desulfurization (FGD) in a scrubber to remove SO_2.
- Flue gas was free of NO_x.
- The equilibrium stage was assumed for both the absorber and the stripping section.
- The Murphree efficiency was assumed to be 1 [30, 31], indicating that the liquid and vapor phases on the tray are well mixed and to obtain results that resembled actual plants.
- The flue gas flow rate and the composition were constant during the simulation

Flue gases from the power plant are assumed to be scrubbed for the removal of SO_2, NO_X, and other impurities before being fed to the bottom of the absorber. Flue gas desulphurization (FGD) is a proven way to reduce SO_2 emissions in pulverized coal combustion (PCC) units and supercritical pulverized coal (SCPC) units [32]. The presence of impurities in the flue gas can lead to an undesirable irreversible reaction with the amine solution and form heat-stable salts that cannot be reclaimed [33]. NO_2 is the main chemical compound from NOx responsible for this irreversible reaction; nevertheless, a recommended desirable level of NO_2 is about 20 ppmv [33, 34]. For SO_X, the installation of the FGD unit is considered to be less cost-effective than the degradation of the solvent when the SO_X level in the flue gas is above 10 ppmv [34].

The flue gas from the power plant is usually cooled in a direct contact cooler to a temperature around 40-50 ^{o}C before being transported to the absorber with the help of a blower [8, 33]. The temperature of the flue gas from a NGCC power plant is around 110-120 ^{o}C and needs to be cooled before it is sent to the absorption system [33].

The flue gas enters at the bottom of the absorber, usually a packed tower, with the absorbent entering counter-currently from the second stage from the top of the column. Cooling water is fed to the absorber top stage to reduce solvent loss; thereafter, the clean gas is vented to the atmosphere. The concentration of MEA in the exit gas is reduced by the makeup water before the pure gas is discharged [35]. The flue gas pressure is increased slightly before being sent to the absorption column to reduce the pressure in the column.

2.2.1.1 Key process simulation inputs

CO_2 capture rates of 90%, 95%, and 99% were considered in both the coal-fired and natural gas-fired power plants. The composition of flue gas from both coal and natural gas used in this study is shown in Table 2.1.

Table 2.1: Flue gas flow rate and composition of flue gases from coal- and natural gas-fired plants.

	Parameters	Unit	PCC	NGCC
	Flow rate	kg/hr	2479931	1615772.89
	Temperature	C	55	50
	Pressure	Bar	1	1
	Plant capacity	MWe	500	180
Composition				
	H_2O	Vol. %	9.41	9.73
	CO_2	Vol. %	12.33	4.41
	N_2	Vol. %	73.49	73.67
	O_2	Vol. %	4.77	11.31
	Ar	Vol. %		0.88

It is necessary to make an informed decision when choosing a particular gas absorption contactor over another for the absorption and stripping columns. The contactor provides a large liquid surface area that comes in contact with the gas phase under conditions supporting mass transfer [36]. Tray columns are useful for large installations, non-foaming liquids, and non-corrosive solvents. However, packed columns are best suited for corrosive fluids, have superior performance, and are widely used for counter-current flows of liquid and gas, as in the case of amine absorption [8, 33, 36]; therefore, in this study, a packed column was considered over trays.

Table 2.2 shows the key design parameters used in the model simulation for absorber and stripper columns.

Table 2.2: Key design parameters of absorber and stripper columns for the simulation model

Parameter	Unit	Absorber	Stripper
CO_2 capture	%	90, 95, 99	90, 95, 99
Number of stages		20	15
Packing type		CMR[3] #2	CMR #2
Packing arrangement		random	random
Packing material		stainless steel	stainless steel
Packing height	m	15	15
Ptop	bar	1	1.42
Press bottom	bar	1.05	1.6
Ttop	C	50.5	100.7
Tbottom	C	67.2	117
Diameter	m	9.8	4.5
Reboiler duty	MW		500

2.2.2 Chemical reaction model

The absorber and stripping units were modelled with Aspen RadFrac [29] in this study with the following equilibrium reactions:

Dissociation of water

---

[3] CMR- Cascade mini rings

$2H_2O \leftrightarrow H_3O^+ + OH^-$ [R1]

Dissociation of CO$_2$

$CO_2 + 2H_2O \leftrightarrow H_3O^+ + HCO_3{}^-$ [R2]

Dissociation of bicarbonate

$HCO_3{}^- + H_2O \leftrightarrow H_3O^+ + CO_3{}^{-2}$ [R3]

Amine protonation

$H_2O + MEA^+ \leftrightarrow H_3O^+ + MEA$ [R4]

Carbamate formation

$MEACOO^- + H_2O \leftrightarrow MEA + HCO_3{}^-$ [R5]

The reversible reaction in a MEA-H$_2$O-CO$_2$ system takes place in an aqueous phase at a given temperature and pressure. During the counter-current flow of the flue gas and amine, the chemical reactions help in the absorption of the CO$_2$ from the flue gas with amine sorbent while the CO$_2$-rich amine is pumped to the stripper and the purge gas is discharged through the upper section of the absorber.

The equilibrium equation is calculated using Eqn. 1. Rate constants are shown in Table 2.3. The rate constants are temperature-dependent with values in ^{0}K.

$$lnK_j = A_j + \frac{Bj}{T} + CjlnT + DjT \qquad (1)$$

Table 2.3: Values of reaction constants for temperature-dependent parameters for equilibrium constants

Parameter	R1	R2	R3	R4	R5
A_j	132.89	231.46	-0.52	-3.038	216.05
B_j	-13445.9	-12092.1	-2545.53	-7008.3	-12431.7
C_j	-22.47	-36.78	0	0	-35.48
D_j	0	0	0	-0.00313	0

Because of structural and turn-down ratio limitations, it is important that the packed column diameter not exceed 12.2 m [37, 38].

The CO_2 production rate is determined through Eqn. 2.

$$CO_2\text{production} = \frac{\text{Mass flow of } CO_2 \text{ in the captured flue gas}}{\text{Mass flow of } CO_2 \text{ in the flue gas stream}} \qquad (2)$$

2.2.3 Process unit operations

CO_2 capture from power plants comprises three main processes: the chemical absorption of CO_2 with MEA, amine regeneration, and CO_2 compression.

2.2.3.1 Absorption unit

The flue gas from point source is assumed to be free from impurities such as NO_x and SO_x after it leaves the FGD unit and to be around 50°C. The absorber was modelled at 1 bar with a pressure drop of 0.05 bar in Aspen Plus V8.8 using the ELECNRTL method and RadFrac [29] with equilibrium stages assumed in both the absorption and stripping columns. The flue gas enters the bottom of the absorber through the blower to compensate for the pressure loss in the absorption process and in an upward direction at a temperature of 53°C [39]. The solvent is fed to the second stage of the column counter-currently while makeup water enters through stage one on the column and helps reduce solvent loss before the cleaned gas is released to the atmosphere.

The lean solvent, with 30 wt% MEA, enters the top second stage counter-currently into the absorption column. The rich amine exits the absorber with a loading of 0.44 mol CO_2/ mol MEA and is pumped to the stripping section. As the flue gas flows to the top of the column, the CO_2 concentration in the flue gas decreases, hence the amine solvent absorbs the CO_2 before it leaves from the bottom of the column. However, the clean gas, scrubbed with the makeup water in the top column, is vented to the atmosphere.

2.2.3.2 Stripping unit

The rich solvent from the absorption unit is pumped to the desorption section through a heat exchanger to a temperature of 100-150°C [40]. The stripper is operated at a pressure of 1.42 bar at the top of the column with a pressure drop of 0.18 bar. This column has the highest energy requirement during the capture process [39]. The required steam is first generated in the power plant and then sent to the reboiler in the column in order to reverse the chemical reaction during the absorption process and therefore strip off the amine solvent used in the absorber while the regenerated amine is recycled back to the absorber.

In order to manage the thermal degradation of amines, the temperature needs to be lowered as well as the liquid hold-up at the bottom of the desorption column. That said, higher temperatures during the stripping of CO_2 together with higher CO_2 absorption heat lower the overall energy requirement in the desorption process [41, 42]. Higher stripper temperatures and pressures help reduce the electricity requirement during CO_2 compression process [30, 43]. In addition, an increase in stripper pressure reduces the size and capital cost of the stripping and compression units [40]. However, in this study, a pressure above atmospheric was considered in order to maintain regeneration conditions [8]. An increase in temperature from 90 to 150 °C reduces about 30% of the equivalent work (W_{eq}). The equivalent work (W_{eq}) comprises reboiler heat duty (Q_i), compression work (W_{comps}), and pump work (W_{pumps}), all of which depend on the reboiler temperature (T_i) and the ambient temperature (T_{sink}) [40]. Consequently, the ideal operating temperature in the regeneration unit is determined by thermal degradation. The equivalent work is estimated using Eqn. 3.

$$Weq = \sum_{i=1}^{n\_reboilers} 0.75 \times Qi \left(\frac{Ti + 5k - T\,sink}{Ti + 5k}\right) + W\,pumps + W\,comps \tag{3}$$

The lean solvent flow rate is assumed to be around 5 times the CO_2 flow rate in the flue gas; the rest of the solvent is about 67% water and 3% CO_2 as the lean solvent contains an insignificant amount of CO_2 [35].

The overhead product, comprises mainly of CO_2 with fractions of water and MEA, passes through a condenser, where the obtained CO_2 product is sent for further processing, primarily in the compression unit. The condensate, partly water and MEA, is refluxed back to the stripper column, while the rest (recycled lean stream) is re-used for scrubbing in the absorber, which helps maintain the water balance in the system [39].

2.2.3.3 CO_2 compression

The captured CO_2 product from flue gas needs to be compressed before being pipelined for storage, enhanced oil recovery (EOR), or use [44]. The main reason for compression is that CO_2 separation from flue gas occurs at low pressure; pressure is elevated through compression, which reduces the volumetric flow and improves transportation. In this study, the compression unit was modeled in Aspen Plus and 4 stages were assumed in keeping with a study by Heischkamp et al. [39], who recommended that the minimum number be 4. There is a wide consensus that energy demand in the entire capture process is greatest in the compression unit, therefore reducing energy consumption in this unit has a significant effect on electricity generation efficiency [39, 45]. Also, it is worth noting that CO_2 compression can be about 8-10% of the total power consumption of the entire process; this is significant and reduces the net efficiency of electricity generation [46]. One way to improve the efficiency in this unit is through inter-stage cooling, which makes the compression process similar to an isothermal process [45]. Table 2.4 shows data input to the inlet of the 4-stage compression unit with an assumed polytropic efficiency[4] of 84% in the first stage and 72% in the final stage of compression. This study saw a 93% decrease in water content from the initial inlet stage to the final stage of the compression unit, resulting in a pure CO_2 almost free of moisture and other gases [45].

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[4] The process of dividing compression path into several small steps while isentropic efficiency is constant along these small steps

Table 2.4: Parameters at the compressor inlet unit of the simulation model

	Coal			Natural Gas		
Capture rate [%]	**90**	**95**	**99**	**90**	**95**	**99**
Mass flow rate of CO_2 [kg/hr] [5]	410000	440000	460000	100000	110000	110000
Inlet temperature [^OC]	35	35	35	35	35	35
Inlet pressure [bar]	1.5	1.5	1.5	2.1	2.1	2.1
Density [kg/m^3]	2.5	2.5	2.5	3.6	3.6	3.6
Moisture content [%]	4	4	3.8	1.2	1.2	1.2
Outlet pressure before pumping [bar]	85	85	85	85	85	85
Outlet temperature [^OC]	50	50	50	35	35	35

The compression unit in Figure 2.2 is a schematic diagram for a 99% capture rate of coal flue gas; however, it was simulated for the different capture rates in coal and natural gas flue gases. The cooling system temperature is assumed in this study to be 35 ^oC. This temperature has a decisive effect on the overall cooling system, as the temperature can enhance the efficacy of the whole process [45]. The permissible pressure drop during the inter-stage process is determined by the formula in Eqn. 4. However, the pressure drop should not be higher than 0.344 bar [45]. Since the reported minimum purity rate of 95% is generally a prerequisite in guaranteeing the mixing ability of the pure product with petroleum, in this study across the different capture rates a purity greater than 99% is obtained with an exit pressure of 85 bar; thereafter, the condensed CO_2 is raised to about 120 bar with the pump for later uses such as EOR [47, 48].

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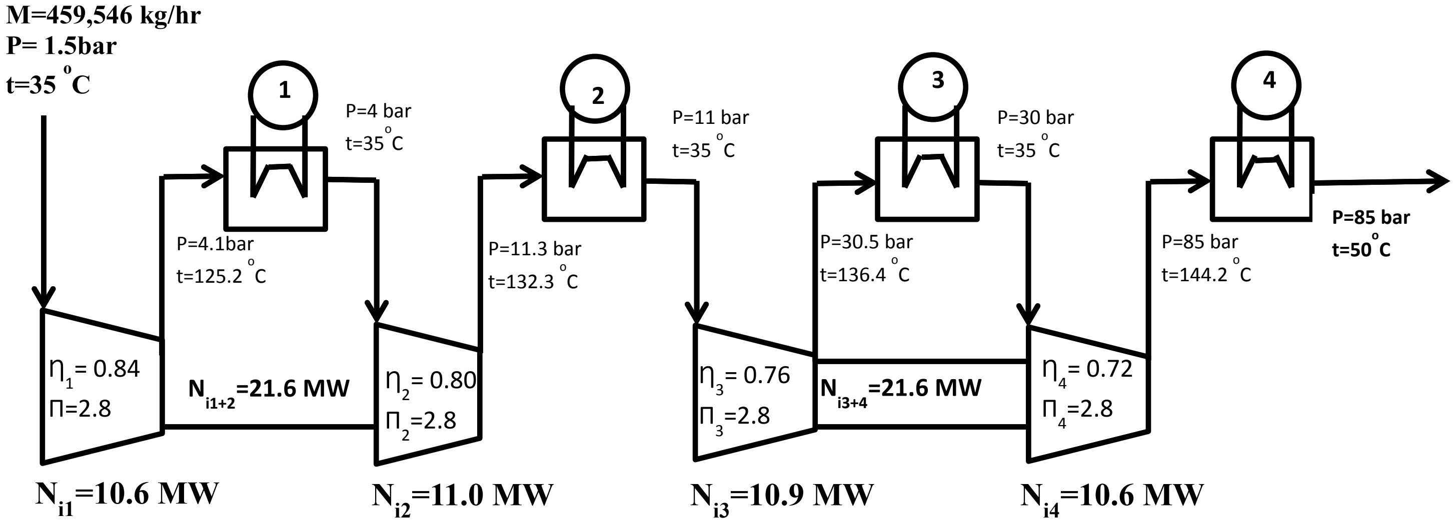
[5] Mass flow of CO_2 at the inlet of the compressor

$$\Delta P = \frac{(14.504 * PCO_2)^{0.7}}{10 * 14.504} bar \tag{4}$$



Symbol	definition
M	Mass flowrate, kg/hr
P	Pressure, bar
t	Temperature, °C
η	Efficiency
Π	Average pressure ratio
N	Compressor internal power, MW

Figure 2.2: The 4-stage compression process with inter-stage coolers as developed in the simulation model

Table 2.5: Values obtained for pressure drop in inter-stage coolers from the simulation model

Inter-stage cooler	ΔP (Bar)
1	0.1211
2	0.2447
3	0.263
4	0.343

The internal power of the compressor shown in Figure 2.2 is 43.14 MW. Table 2.5 shows the values obtained in the 4-stage compression process. The individual permissible limit of the pressure drop calculated using Eqn. 4 should not exceed 0.344 bar. Beyond this, permissible pressure drop results in CO_2 velocity increase, thereby increasing the heat exchange, which ultimately reduces the required heat exchange surface area [46].

Accurate selection of the structure, the surface area and the configuration of inter-stage exchangers play a key role in determining the cost effectiveness of the compression system; for example, it is imperative that the gas stream pressure drop be minimized [46]. Other challenges include the corrosiveness of the pure CO_2, hence stainless steel is preferable in the design in order to avoid corrosion hazard.

2.3 Techno-economic analysis

In order to determine the cost of producing pure CO_2 in \$/tonne, several cost analysis parameters were considered and are discussed in this section.

2.3.1 Cost model parameters

A base process model was simulated in Aspen Plus for flue gases from both a 180 MWe natural gas-fired power plant and a 500 MWe coal-fired power plant and the results loaded to the Aspen Process Economic Analyzer (APEA) for the techno-economic analysis. The APEA modelling tool simulates the various process equipment connected by mass, energy, and work streams [49]. APEA also helps determine equipment cost. To compare our CO_2 production costs with those from other studies, we scaled up the simulated model for NGCC to 555 MW_e using inbuilt scale up process in APEA [50]. In the APEA platform, the equipment was mapped and sized based on their design parameters. Alberta-specific rates for labor and supervisor salaries were used in the cost model since we assumed that the plant is located in Western Canada. After sizing equipment in APEA, we estimated the costs of the various equipment, i.e., the absorber, regenerator, CO_2 compressors, pumps, heat exchanger, flue gas blower. The total project investment (TPI) is the sum of the direct equipment costs and the indirect costs calculated as fractions of the total purchased equipment cost (TPEC) [19] as shown in Table 2.6. The capital cost was estimated based on the total equipment purchase after the mapping of the equipment. The same approach has been used in studies by Oyedun and Kumar [51], Shahrukh et al. [52], Agbor et al. [53] and Kumar et al. [49] based on the studies by Peters et al. [54].

Table 2.6: Plant capital investment cost factors [54].

Total purchased equipment cost (TPEC)	100% TPEC
Total installed cost (TIC)	302% TPEC
Indirect cost (IC)	89% TPEC
Total direct and indirect cost (TDIC)	TIC + IC
Contingency	20% TDIC
Fixed capital investment (FCI)	TDIC + Contingency
Location factor (LF)	10% FCI
Total project investment (TPI)	FCI + LF

The operating cost includes utility cost, labor and maintenance costs, operating charges (25% of labor and maintenance costs), plant overhead (50% of operating and maintenance costs), and general and administrative costs (G & A) [55]. Other variable operating and maintenance costs include MEA solvent cost and water cost. The cost of MEA solvent was calculated using Eqn. 5 adopted from Rao et al. [56]:

$$\mathrm{VOM}_{MEA} = \mathrm{M}_{MEA,makeup} \times \mathrm{UC}_{MEA} \times \mathrm{HPY} \tag{5}$$

where VOM_{MEA} = Cost of MEA solvent (\$/tonne solvent), $\mathrm{M}_{\mathrm{MEA,\ makeup}}$ = Cost of MEA makeup (\$/tonne makeup solvent), UC_{MEA} = unit cost of MEA (\$/tonne MEA), HPY = Hours per year.

In the plant operation, 6 operators per shift were assumed with 1 supervisor for both base plants considered in this study close to what was reported by Parsons et al. [57]. Unless otherwise stated, the 2016 USD was used; all values were converted to the cost base year and currency. All cost estimates were converted to 2016 using the Consumer Price Index (CPI) formula in Eqn. 6:

$$\frac{\text{CPI 2016}}{\text{CPI Year X}^{6}} \times \text{Year X (USD)} = \text{2016 USD} \tag{6}$$

Table 2.7 shows in detail the parameters used in the economic analysis. The capture plant has an economic life of 20 years in addition to a 3-year design and construction period with 20%, 35%, and 45% allocated to the project capital cost for Years 1, 2, and 3, respectively [52, 53]. The product value, which is the capture cost in \$/tonne CO_2, is obtained from the discounted cash flow (DCF) spreadsheet developed for the various capture rates. An IRR of 10% was considered with a plant capacity factor of 0.85 [52, 53]. The cost of electricity is \$0.04 /kWh according to rates in Edmonton, Alberta in 2016 [58] and MEA reagent cost is \$3715 /tonne CO_2 [59].

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[6] Year X is the reference year

Table 2.7 Techno-economic analysis modelling input

Parameters	**Unit**	**Value**	**References/ comments**
Plant life	Y	20	Assumed
Cost year basis[7]	USD	2016	
Design and construction period			[52, 53]
Year 1	%	20	
Year 2	%	35	
Year 3	%	45	
Plant capacity factors			[52, 53]
Year 1	%	70	
Year 2	%	80	
Year 3 and onward	%	85	
Maintenance cost	$	3% of TPI	
Operating charges	$	25% of operating labor cost	
Operating cost subtotal	$	Sum of all operating costs including raw material and utility costs	
G & A	$	8% of operating cost subtotal	[51]
Internal rate of return (IRR)	%	10	
Unit cost of electricity	$/kWh	0.04	[58]
Operating hours per year	hr/yr	8000	
MEA reagent cost	$/tonne	3715[8]	[59]
MEA losses estimation	kg MEA/tonne CO_2	1.5	[56]

---

[7] 2016 USD unless otherwise stated

[8] Converted to 2016 USD

2.4 Results and discussion

2.4.1 Effects of operating parameters (process model results)

Several parameters, including the number of stages in the absorber column and the absorber and stripper temperatures, were varied to study their effect on the thermal energy requirement of the capturing process. The absorber is a significant part of the overall capture cost; therefore, any parameter that influences the absorber performance has an impact on overall costs. As shown in Figure 2.3, an increase in the number of stages increases the capture rate with a corresponding decrease in thermal energy requirement. Beyond 20 stages, the reduction in thermal energy is insignificant, therefore 20 stages were considered for this study.

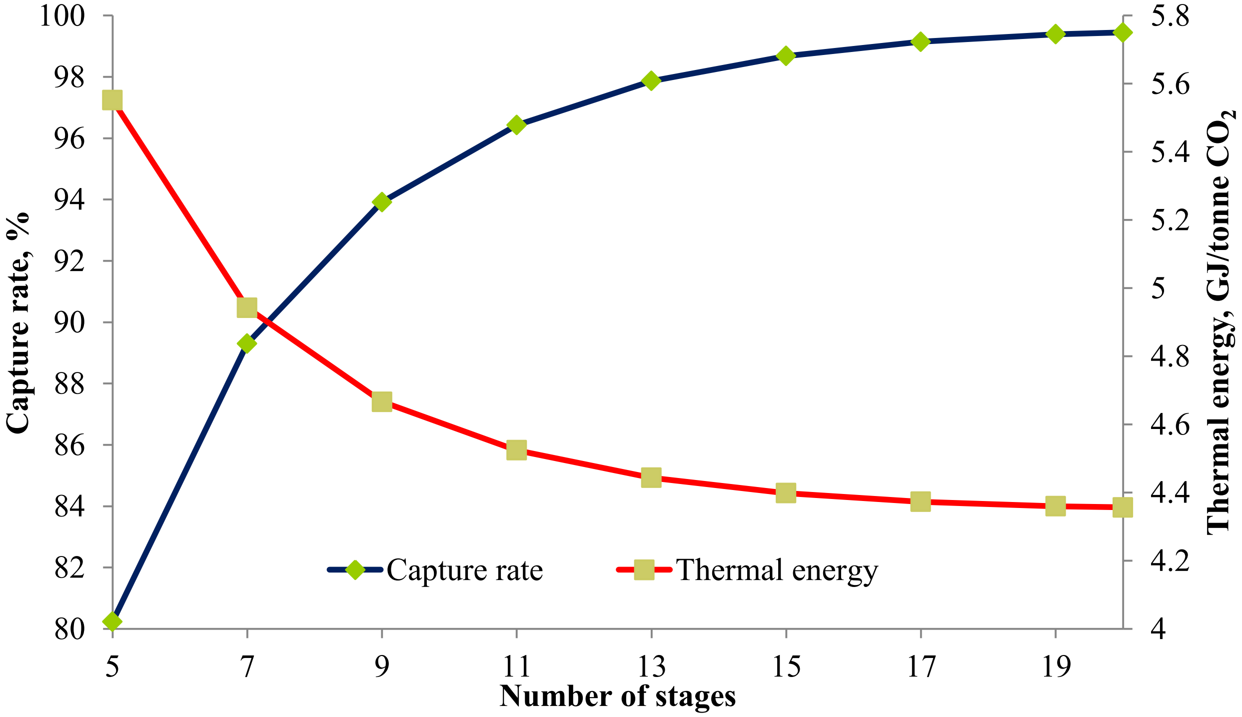

Figure 2.3: Effect of the number of stages on capture rate and thermal energy

Figure 2.4 and 2.5 show the effects of stripper pressure and absorber temperature, respectively, on the capturing of CO_2 from coal plants. An increase in stripper pressure significantly decreases the regeneration energy even though the reboiler temperature increases, as shown in Figure 2.4. Likewise, thermal energy use decreases with an increase in absorber temperature, which could increase CO_2 purity and, with it, production cost. As reported by Goto et al.[60], higher CO_2 purities require higher power requirement. Their results show that a decrease of CO_2 purity target from 99% to 95% saw a 10% decrease in energy requirement. Nevertheless, it is expected that higher amine degradation rates and corrosion will occur at these elevated pressures and temperatures [8], therefore a stripper pressure of 1.8 bar with a reboiler temperature of 126 ^{0}C was used in the model.

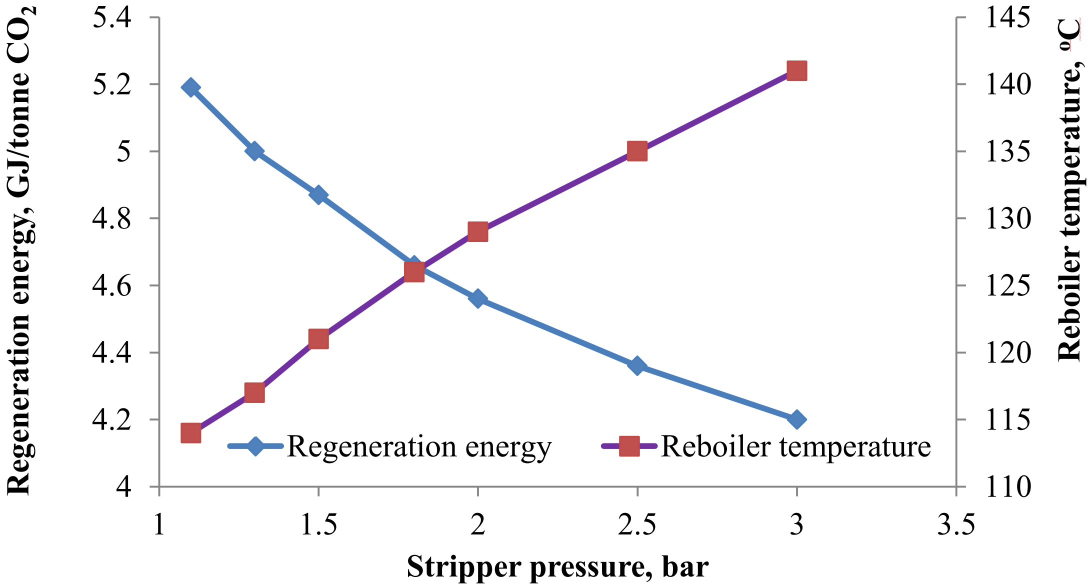

Figure 2.4: Effect of stripper pressure on regeneration energy and reboiler temperature

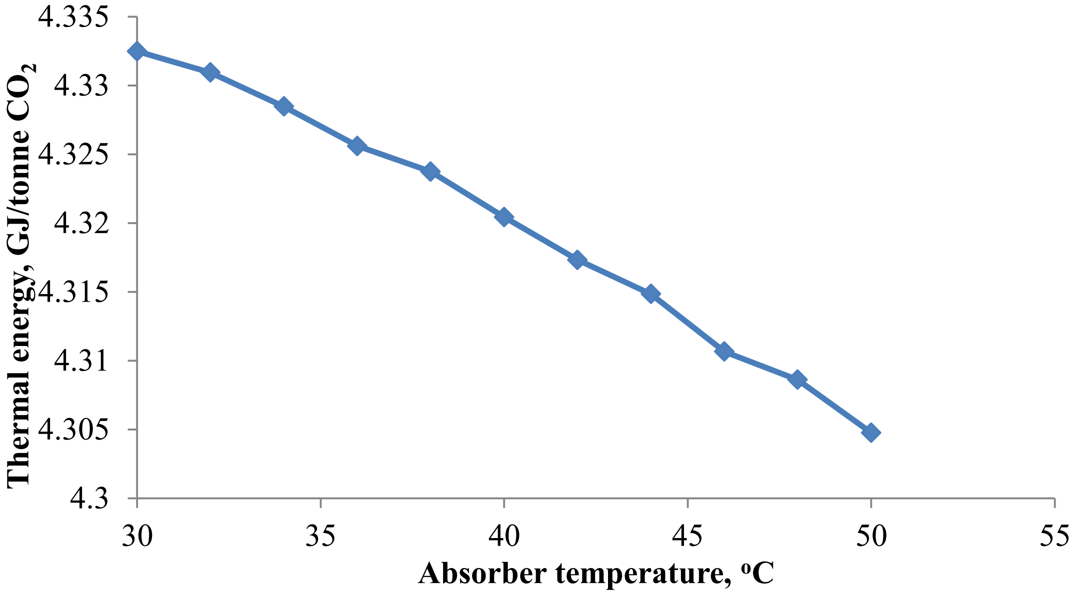

Figure 2.5: Effect of absorber temperature on thermal energy

2.4.2 Purity of captured CO_2 streams

The captured CO_2 stream results for flue gas from coal and natural gas plants at different capturing rates are presented in Table 2.8. The main focus of this study was the capture of CO_2 at a production rate of 90% and higher with a purity high enough for commercial use. The final product purities after compression are 99.7% and 99.6% for coal and natural gas, respectively. The purity level for CO_2 products should be above 99% to avoid corrosion in pipes during transportation, and the water level should be low [56]. Rao et al. also noted that other impurities such as nitrogen should be reduced as they may pose threats during compression and CO_2 liquefaction [56]; moreover, the impurities could have a negative effect on the performance and cost of the capture system [19].

Table 2.8: Purity of captured CO_2 streams for both coal and natural gas plants

Capture Products	90%	95%	99%
Coal			
CO_2	0.9	0.95	0.99
H_2O	0.000314	0.00033	0.0003
O_2	0.000025	0.000027	0.000027
N_2	0.000014	0.000014	0.000014
Purity, %	99.7	99.7	99.7
Natural gas			
CO_2	0.9	0.95	0.99
H_2O	0.908095	0.952248	0.98597
O_2	0.0000359	0.0000359	0.00000359
N_2	0.0000194	0.0000194	0.0000194
Purity,%	99.6	99.6	99.6
Temperature, ^oC	50	50	50
Pressure, bar	85	85	85
Compression stages	4	4	4

2.4.3 Capital and operating costs

Figure 2.6 shows the equipment cost distribution at a 90% capture rate for coal and natural gas flue gas. The cost distribution is similar to the 95% and 99% capture rates. The equipment that contributes most to the overall equipment costs are the absorber, stripper unit, compression unit, and heat exchangers. These equipment makes up more than 70% of the overall capital expenditures in both the natural gas combined cycle and coal-fired power plant cost analyses, similar to what has been reported in other studies [27, 61, 62]. In general, the absorption unit is the most expensive piece of equipment in both pulverized coal (PC) and natural gas combined cycle (NGCC) plants. Also, the compression unit is particularly necessary in reducing the amount of moisture in the stream to avoid corrosion during pipeline transportation [33].

For MEA absorption processes, the cost of the absorber is directly linked to the flue gas flow rate, as this equipment usually handles a large volumetric flue gas flow rate and is also the largest piece of equipment in a capture plant [8, 19]. Furthermore, it is assumed in this study that the absorber and stripper are made of steel, which contributes to their high cost even though steel mitigates the corrosive nature of the flue gas.

A natural gas combined cycle plant has a lower CO_2 concentration in its flue gas than a coal-fired power plant and therefore has a higher energy demand during the absorption and desorption processes. The effect is reflected on the overall cost ($/tonne), which is evident in the higher cost of CO_2 production from a natural gas-fired plant than a coal-fired power plant.

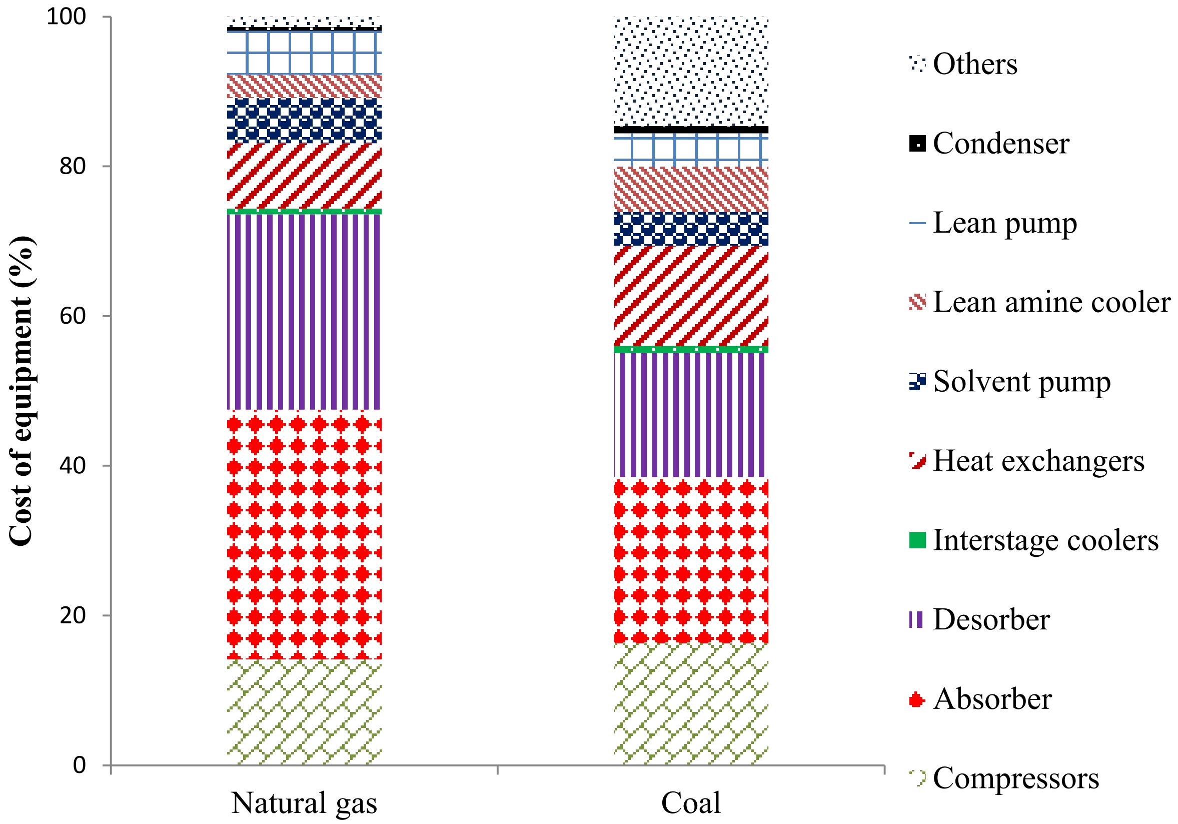

Figure 2.6: Percentage breakdown of equipment cost for a 90% capture rate.

2.4.4 Capture cost (CO_2 production cost)

As indicated earlier, capture cost is simply the cost of CO_2 capture in a reference plant. Several parameters, such as temperature, absorber pressure, and the number of stages, were varied to determine an optimum capture cost. An amine separation unit requires high energy for solvent regeneration prior to the solvent removal in the column [19]. So for the regeneration unit, an optimum pressure of 1.8 bar and temperature of 126 ^{o}C were used in the model. In this study, the cost of capture was determined at different capture rates with a purity of 99% or higher since our main objective was to capture CO_2 for use. The capture rate assumed in most studies is usually 90%. In order to compare the production cost of CO_2 in this study with other studies, a 90% capture rate was chosen in this study and is discussed in this section below.

Table 2.9: Summary of cost estimates for different capture rate scenarios

	Coal (500 MWe)			Natural Gas (555 MWe)		
Cost parameters	90%	95%	99%	90%	95%	99%
Total purchase equipment cost (M$)	163.1	178.9	190.9	150	176.3	193.6
Total project investment (M$)	842.1	923.4	985.3	774.3	901	999.4
Operating cost (M$)	36.1	38.5	40.4	23.2	38.1	40.8
Cost of capture ($/tonne CO_2)	60.65	61.57	62.16	79.47	84.44	87.07

Table 2.9 shows the main cost parameters incurred for the cost of CO_2 capture for flue gas from both coal-fired and NGCC plants. The operating cost comprises operating and maintenance (O&M) costs assuming 6 operators per shift and 1 supervisor. Since the plant's assumed location is Western Canada, the wage rates for this region are used for the model. The total project investment cost is estimated to be M$ 842.1 with a production cost of $60.65 /tonne CO_2 for flue gas from a PC plant for a 90% capture rate. On the other hand, the total investment cost for flue gas from a NGCC plant is M$ 774.3 with a production cost of $79.47 /tonne CO_2 for flue gas for a 90% capture rate. The NGCC power plant's capture cost is higher than a PC's because the former has low CO_2 content in the flue gas and a higher energy intensity [63]. Flue gas from PC plants has a higher CO_2 content, which translates to a lower energy requirement than a NGCC plant.

Figure 2.7 shows the cost breakdown of different parameters in capture rate scenarios of 90%, 95%, and 99%. The CO_2 production costs are lower from coal flue gas than from natural gas for all capture rates. Different capture costs have been reported in literature. Rubin et al., for instance, reported costs of CO_2 avoided for a NGCC plant as $49.38-98.76 /tonne CO_2 and for a coal plant as \$38.70-68.07 /tonne CO_2 for a net plant output of 500 MW [64]. In another study, the cost of CO_2 avoided was \$66.11 /tonne CO_2 for a plant capacity of 600 MW [65]. Klemes et al. link capture cost with plant capacity. For net outputs of 300-900 MW, the cost of CO_2 avoided is from \$82-57 /tonne; for capacities of 900-1500 MW, the CO_2 capture rate is 57-42 \$/tonne and for capacities of 1500-2000 MW the capture cost is \$42-38 /tonne CO_2 [66]. The cost parameters outlined earlier include the cost of CO_2 avoided, which is often accompanied by a penalty for the emissions of CO_2 from burning fossil fuels and has a corresponding impact on consumer electricity pricing [15, 16]. However, our aim is to estimate the cost of producing pure CO_2 as a raw material for reuse. Nevertheless, a trend is noticed with respect to the effect of plant size and associated costs. As plant capacity increases, capture cost decreases. This is true of an NGCC with a 90% capture rate and plant capacity of 180 MW, which results in a production cost of \$104.19 /tonne CO_2, while the production cost is \$79.47 /tonne CO_2 from a 555 MW plant capacity, a drop of approximately 24%, as shown in Figure 2.7.

It is important to note that the amount of CO_2 avoided compared to the reference case is usually lower than the captured amount; for this reason, it is widely believed that the avoided cost is usually higher than the cost of CO_2 captured [16].

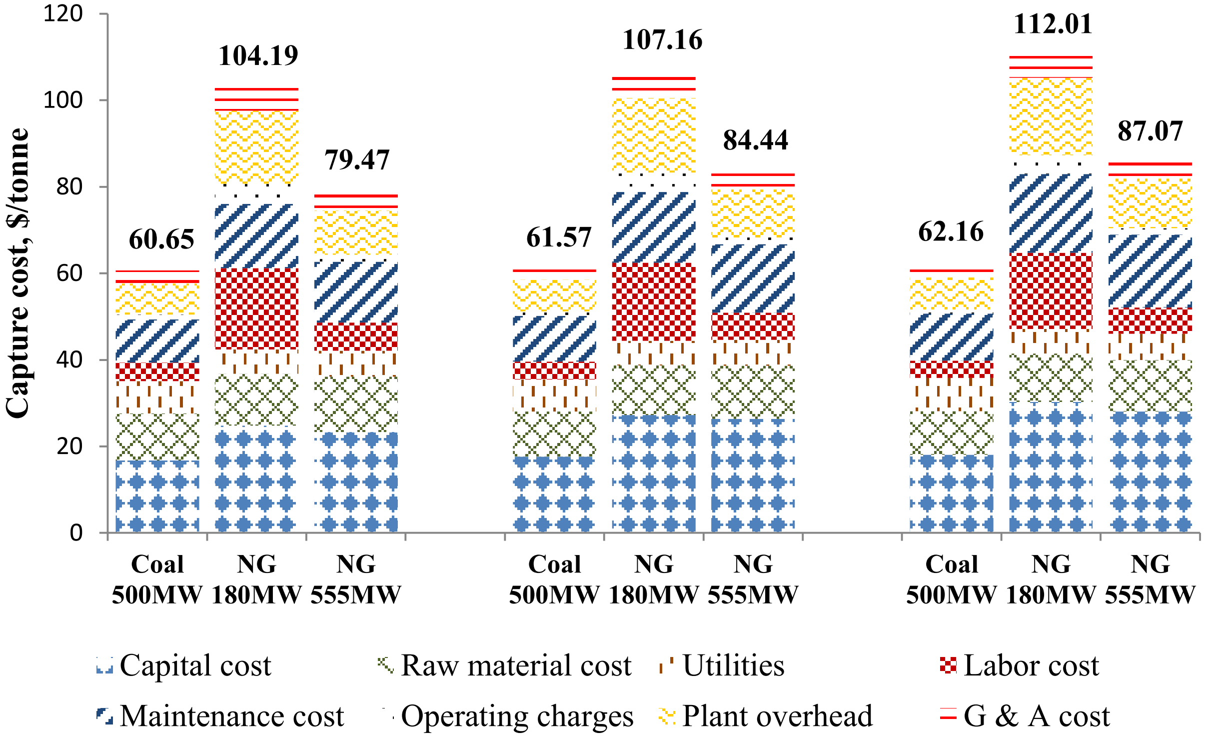

Figure 2.7: Capture costs for different capture rates from coal power and natural gas plants

2.4.5 Comparative studies

Table 2.10 lists various studies specifically on NGCC plants with a flue gas CO_2 concentration less than 5 mol%, an amine concentration of 30 wt%, and a 90% capture rate [18, 21, 67, 68] . The capture cost found by Sipocz and Tobiesen is 103.13 \$/tonne CO_2 [21], and in this study the cost is 79.47 \$/tonne CO_2. One reason for this disparity is differences in equipment sizing. Sipocz and Tobiesen report the absorber to be 26.9 m high and 9.13 m in diameter and the stripper to be 23.5 m high and 5.5 m in diameter. Therefore, in our equipment design, we used the optimum height of 15 m and absorber diameter of 9.8 m, which resulted in a pure CO_2 cost of

\$79.47 /tonne CO_2 compared to 103.13 \$/tonne CO_2. The absorber and stripper are a significant part of the equipment cost, as shown in Figure 2.6.

Several studies report that the equipment that makes up more than 50% of the capital cost are the absorber, stripper, compression unit, and heat exchanger [61, 62, 66, 69]; we came to the same conclusion in our study. Hendriks determined capture cost for a plant capacity of 600 MWe to be 66.11\$/tonne CO_2 [65]. According to Bogner et al. [70], capture cost shows the viability of a CO_2 capture system in reference to the market price for CO_2, considered an industrial commodity. In other words, CO_2 as a commodity serves as a raw material to other industries, for example in enhanced oil recovery (EOR). The cost of CO_2 in EOR operations has been reported to be about \$40-65/tonne CO_2 [71, 72]. DTI UK reported a range for CCU, particularly for EOR, to be around \$65.74-82.16/tonne CO_2 [73], which is on par with the cost of CO_2 capture obtained in this study.

Table 2.10: Comparison of plant performance results with other studies

Parameter	Kvamsdal et al. [67]	Sipöcz and Tobiesen [21]	Biliyok et al. [18]	Agbonghae et al. [68]	This study
Power plant size, MWe	450	440	440	450	555
Flue gas flow rate, kg/s	1045.6	639.61	693.6	725	1382.3
CO_2 concentration, mol%	3.5	4.2	3.996	4	4.41
CO_2 capture level, %	90	90	90	90	90
MEA concentration, wt%	30	30	30	30	30
Liquid/gas ratio, g/g	0.87	0.68	1.04	0.96	3.12
Lean loading, mol CO_2/mol MEA	0.216	0.132	0.234	0.2	0.12
Rich loading, mol CO_2/mol MEA	0.47	0.473	0.495	0.483	0.43
Specific duty, GJ/tonne CO_2	3.77	3.97	4.003	3.96	4.62
Cost of capture (2016 cost)	-	103.13	98.58	-	79.47

2.4.6 Sensitivity Study

The effects of parameters on CO_2 capture cost with a base case of a 90% capture rate for both PC and NGCC were looked into. We varied capital cost, operating and maintenance (O&M) cost by ±30%, similar to the work of Kolstad and Young [74], internal rate of return (IRR) is varied from 5% to 20% [55] while ±20% was assumed for raw material, operating charges, labor cost, utilities cost, plant overhead cost, , and general and administrative (G&A) expenses. As shown in

Figure 2.8, the parameters that have the most effect on the output are capital cost and IRR, followed closely by O&M and labor costs. The product value (PV) ranges from \$52.36 /tonne CO_2 to \$68.95 /tonne CO_2 when capital cost is changed by ±30%, for a PC plant and \$69.51 /tonne CO_2 to \$89.44 /tonne CO_2 for a NGCC plant. It is important to note that a decrease in the diameter of the absorber and stripper columns, combined with a decrease in process thermal energy requirement, could lower the PV considerably. Similar trends were noticed for capture rate scenarios of 95% and 99%.

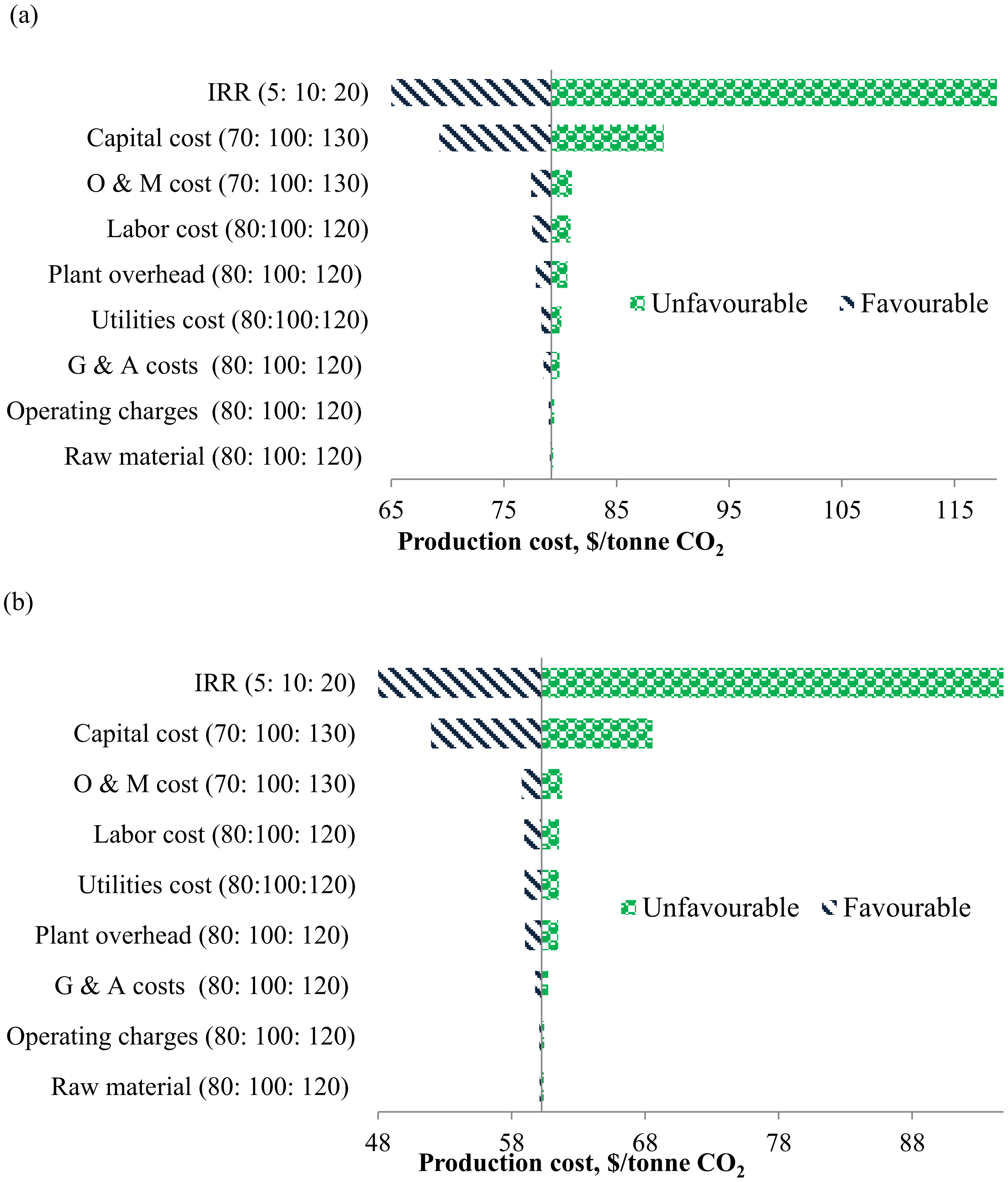

Figure 2.8: Sensitivity analysis of major parameters in 90% CO_2 capture in the (a) NGCC scenario and (b) PC scenario

2.4.7 Uncertainty analysis

During model simulations, some assumptions were made that could have an impact on the overall cost estimates. Though efforts were made to achieve accurate results, the effects of imperfect data on cost analysis cannot be ignored. Therefore it is essential to factor in the various uncertainties during cost estimations. This analysis enables us to see the impact of uncertainties on the PV cost parameters on the different flue gas sources. The Monte Carlo simulation method was used with a model risk tool, an Excel-based software with about 10,000 iterations run during the simulation [75]. In order to carry out uncertainty analysis, we identified the cost parameters that have significant impact on the overall cost and examined uncertainty on these key parameters. However, there is a need to provide some level of uncertainties on the cost of producing pure CO_2.

Figure 2.9 is the graphical representation of the uncertainty analysis results. The simulation results show that the cost of CO_2 capture at a 95% confidence level for PC is \$60.61 ± 3.62 /tonne CO_2; for NGCC, the cost ranges are \$79.25 ± 5.12 /tonne CO_2.

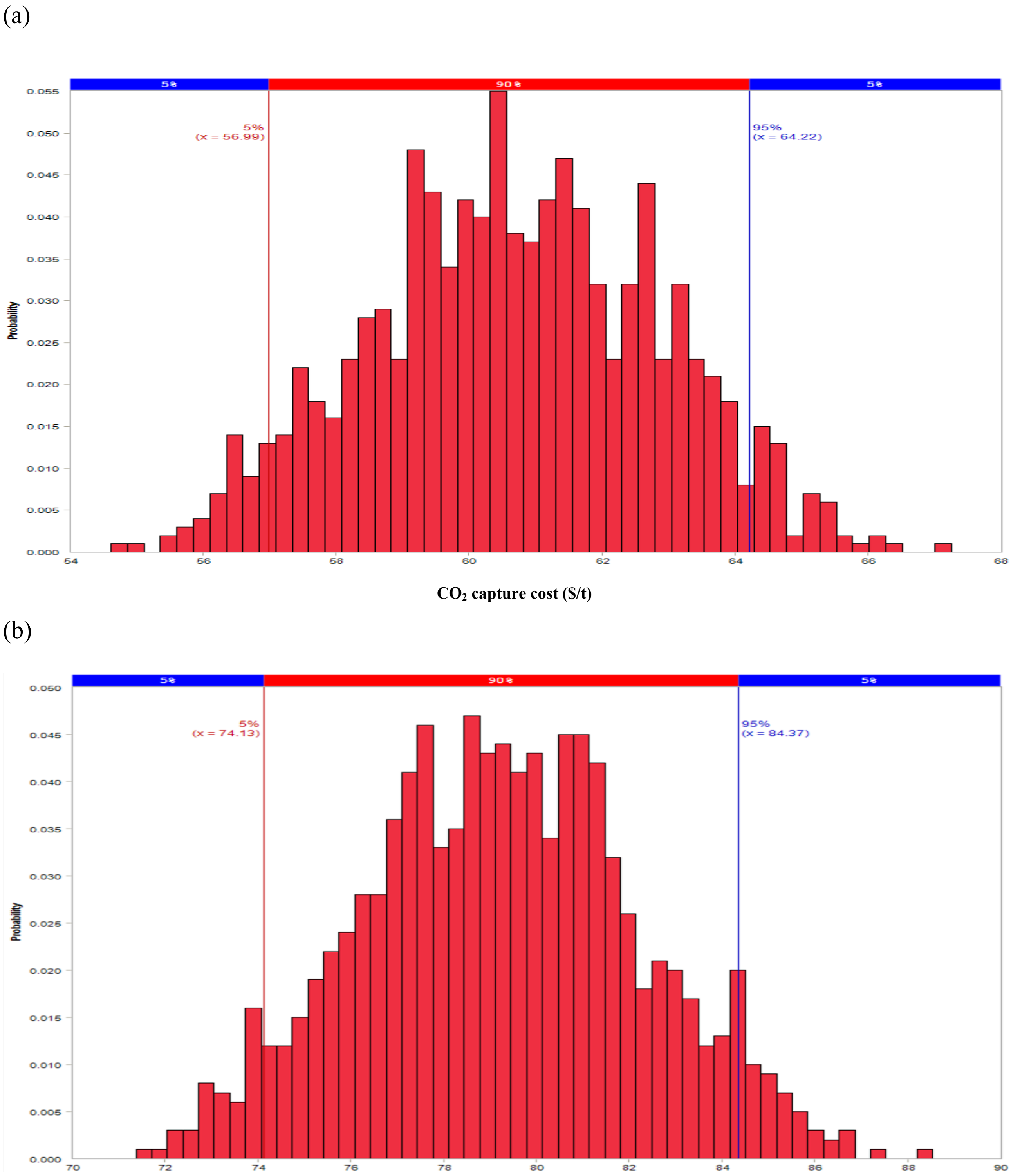

Figure 2.9: Monte Carlo uncertainty analysis results for 90% CO_2 capture cost (a) PC and (b) NGCC plants

2.5 Conclusion

The use of carbon capture and storage for carbon emissions reduction has received great attention over the years. In this study, a detailed techno-economic assessment was conducted through development of data intensive process simulation models to assess the production of pure CO_2 from coal and natural gas power plants with the goal of using CO_2 as a feedstock. The analysis was conducted for the cost of CO_2 production considering capture rates of 90%, 95%, and 99% from both flue coal and NG gases via the MEA capture process. Since most applications of CO_2, including EOR, require high purity, a purity greater than 99% was obtained for both PC and NGCC. The results show that the PC capture costs for all three capture rates are somewhat lower than those from NGCC. This is attributed to lower CO_2 concentration (usually 3%) in flue gas from NGCC, which results in a higher energy intensity in NGCC than the PC plant. In order to obtain pure CO_2 in optimum operating conditions, the effect of changing some parameter values was studied. It was observed that an increase in the number of stages increases in the production rate and decreases thermal energy. However, beyond 20 stages, there was no further decrease in thermal energy requirement. Likewise, regeneration energy decreases with an increase in stripper pressure, which increases the reboiler temperature. The optimum pressure used in this model was 1.8 bar, corresponding to a reboiler temperature of 126 ^{0}C in order to avoid amine degradation. The model results show that at a 90% capture rate, the cost of CO_2 capture is \$60.61 ± 3.62 /tonne CO_2 for flue gas from coal-fired plants and \$79.25 ± 5.12 /tonne CO_2 for flue gas from NGCC plants. The sensitivity analysis for both scenarios shows that the most important cost parameters considered are capital cost and IRR, followed closely by O&M and labor costs. The results of the study will help decision makers across different jurisdictions make investment decisions in the area of CO_2 use. The results will also be of interest to those who invest in chemical processes that use pure CO_2 as feedstock.

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Chapter 3: Assessment of cost of production of pure CO_2 from coal plant flue gas using ammonia separation processes

3.1 Introduction

Fossil fuels are the most widely used source of energy and its combustion contributes significantly to overall greenhouse gas (GHG) emissions worldwide [1]. In order to meet global climate targets, thereby avoiding global warming, a concerted effort to reduce these emissions is needed [1]. Several approaches for reducing overall GHGs, such as fuel switching, including the use of nuclear power and renewable energy, and improving the energy efficiency of the combustion process, have been suggested [2]. Global electricity production in 2014 was 80 EJ, 66% of which were generated from fossil fuels [3]. It is projected that by 2095, electricity generation will be 319 EJ, with electricity from fossil fuels accounting for 76% [4]. Carbon capture and storage has been suggested by researchers as one pathway to mitigate anthropogenic CO_2 emissions [5, 6]. Post-combustion capture has received great attention over the years and is considered a mature technology [7].

Post-combustion capture refers to capturing CO_2 from flue gases (from coal- and natural gas-fired plants, for example) using liquid solvents such as amine and ammonia. Post-combustion technology can be retrofitted into an existing plant or built with a new plant. Amine separation (especially using monoethanolamine [MEA] as a solvent), is the most widely studied technology and has gained significant acceptance through the well-established commercialization status it has held for almost a decade [8, 9]. It also has high CO_2 capture efficiency for sequestration [10]. However, there are limitations to amine separation: high energy consumption, high equipment corrosion, amine degradation by SO_2, NO_2, HCl, etc., and large equipment size requirement [9, 11, 12]. 70% of the operating costs are attributed to regeneration energy during the capture process [13]. These limitations indicate the need for a more energy-efficient, less corrosive, and more cost-effective liquid solvent for carbon capture processes, and ammonia is seen as a better alternative to amine.

The use of aqueous ammonia solvent for CO_2 absorption has received great attention over the years. Several authors discussed the numerous advantages of the use of ammonia solvent over MEA in the capture processes, in particular its lower regeneration energy requirement [10, 14, 15]. The ammonia-based process is also preferred because of its ability to withstand the decomposing reactions/effects of SOx and NOx present in the flue gases [6]. Other benefits include a reduced steam load, the ability to increase CO_2 carrying capacity, thereby lowering the amount of steam load, and the ability to operate at high pressure in the CO_2 capture process, which helps reduce product gas compression costs [16]. In addition, the ammonia solvent is used to capture acid gases associated with the flue gases, such as SO_x and NOx, during the CO_2 capture process [12]. Another economic benefit of using ammonia is the solvent cost; it is an estimated \$263/tonne (\$0.29/kg), lower than the amine cost of \$1360 /tonne (\$1.5/kg).

However, there are drawbacks with ammonia solvent such as ammonia slip, increased equipment costs and auxiliary loads, and slower reaction rates during absorption, which could require the use of large absorption columns [17-20]. Ammonia's volatile nature leads to large amounts of solvent exiting the absorption column with the treated flue gases. Since exposure to high concentrations of ammonia is toxic to humans, care must be taken to lower the chemical substance. In order to control ammonia slip, it is imperative to reduce the ammonia concentration in the solvent, lower the absorption temperature, and have high CO_2 loading [20].

However, the drawback with reduced ammonia slip is a high chilling duty and difficulty handling the resultant slurry [21].

Several studies have been conducted on the use of ammonia-based solvent for the CO_2 capture process. Versteeg and Rubin [22] evaluated the performance and cost of CO_2 avoided using ammonia-based solvent on flue gases from a pulverized coal (PC) plant. They noted that a lower temperature helps reduce the vapor pressure of ammonia above the absorber, thus decreasing the amount of ammonia solvent in the exit gas. Also, Darde et al. [23] studied CO_2 capture process using chilled ammonia as a solvent with absorption temperatures of 0-20 ^{o}C. They developed their process in an equilibrium-based model of the CO_2-NH_3-H_2O system and concluded that the ammonia process uses significantly less energy than the amine process.

In terms of economic analysis, Li et al. [24] conducted a techno-economic assessment of advanced aqueous ammonia-based post-combustion capture with a plant capacity of 650 MW. The cost of CO_2 avoided in the ammonia process was lower than in the MEA process, i.e., US$ 67 /tonne CO_2 vs. US$ 86.4 /tonne CO_2. One of the reasons given for the lower cost is the lower energy requirement. Versteeg and Rubin's [22] performance cost analysis highlighted the similarity in the capture cost of CO_2 avoided using ammonia-based (US$ 73.2 /tonne CO_2) and amine-based technology (US$ 72.2 /tonne CO_2). They also found that the high NH_3 concentration results in high solids precipitation and NH_3 slip. The formation of solids could lead to operational challenges and subsequently equipment blockages, as reported by Alstom at We Energies' pilot plant, and risk plugging in a packed column [25, 26]. Jilvero et al. [20] did a detailed economic assessment on the control of ammonia emissions and concluded that ammonia slip control poses a challenge for the success of ammonia capture process. For example, a decrease in flue gas CO_2 concentration to 5% with a high cooling temperature (25 ^oC) increases the utility cost [20].

CCS is considered a viable technology that could reduce CO_2 emissions [27]. However, the technical and economic shortcomings have affected its large-scale deployment [28, 29]. Therefore, CCU is an attractive alternative in GHG mitigation and in making use of the produced CO_2 in commercial products. In other words, it could serve as revenue to offset the cost of capture and storage. To date, most studies on carbon capture separation processes have focused on the CO_2 avoided cost calculation and not the cost of producing CO_2 from the process. The CO_2 production cost estimate is important for the economic considerations of the CCU, and beyond carbon capture and storage (CCS), carbon capture and use (CCU) is considered the way forward to capturing and re-using CO_2 for commercialization. In this study, the emphasis is on the production cost of pure CO_2 from coal-fired power plant flue gas. While a number of studies have calculated the economics of capturing CO_2 through ammonia, there are few or limited studies on the techno-economic analyses of the production cost of pure CO_2 from flue gases from coal-fired power plants using ammonia solvents. Therefore, in this study, a detailed, economic assessment of producing pure CO_2 using ammonia is conducted. The specific objectives are:

- To develop a base process simulation model for producing pure CO_2 from coal-fired power plants.
- To conduct a detailed techno-economic analysis to determine the cost of producing pure CO_2 from coal-fired power plants in \$/tonne using ammonia solvents.
- To identify the influence of process and cost parameters on the production cost of CO_2.
- To perform both sensitivity and uncertainty analyses on the cost parameters.

3.2 Method

This study conducts a techno-economic analysis of producing pure CO_2 from power plant flue gas using ammonia as solvent. The capture process use a direct contact cooling unit (DCC), a capture unit, and a CO_2 compression unit. The power plant flue gas is cooled in the DCC unit to the desired temperature before being fed to the two absorption columns, where the CO_2 is absorbed by the ammonia solvent as shown in Figure 3.1. The pure gas, after exiting the column, is washed to remove the ammonia concentration to an allowable amount before being discharged to the atmosphere. Thereafter, the rich solvent is pumped to the stripper column.

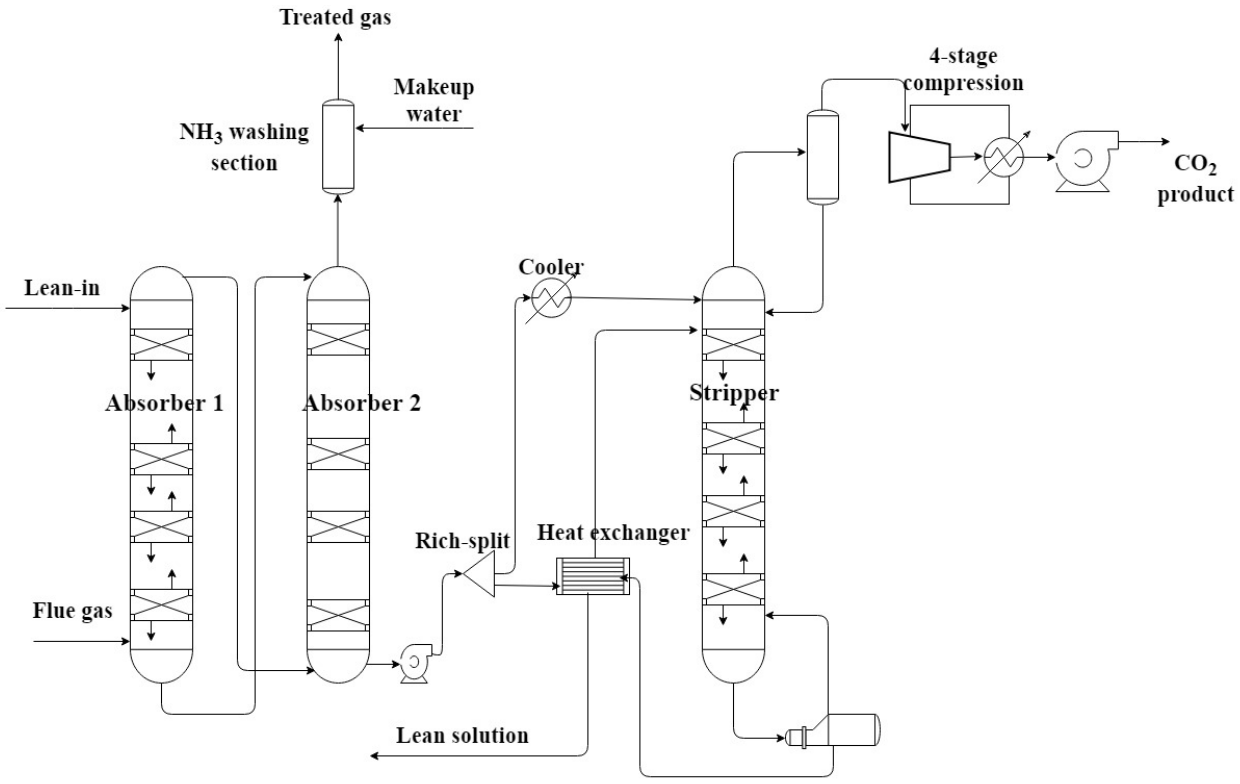

Figure 3.1: Schematic flowsheet of NH_3-based CO_2 production

The flue gas data used in the process model is from a coal-fired power plant with a capacity of 500 MW. The chilled ammonia process (CAP) has received significant attention over the years in CO_2 emissions reduction and was, therefore, applied in this research. Apart from the environmental concerns of ammonia volatility, solids formation in the absorber poses a great challenge. Both high ammonia solvent concentration and high CO_2 loading can lead to solids formation, as others have found [10, 17, 22]. To avoid solids formation and a potential plant shutdown [30], we have decreased the ammonia concentration, as suggested by Sutter et al. [31].

3.2.1 Process model description

The CO_2 from the flue gas capture model was developed in Aspen Plus V8.8 [32]. Table 3.1 shows the flue gas flow rate and composition used in the process model for a 500 MW plant capacity coal power plant. The flue gas data and process conditions were derived from a research carried out by Fisher et al., [33]. The flue gas from the power plant usually comes with an elevated temperature; it was cooled down to 50^oC and assumed to have gone through the flue gas desulphurization (FGD) unit to remove the SO_X and NO_X components.

A two-stage absorption configuration was assumed in this study, as it is reported to reduce ammonia slip by 50% compared to a single-stage column [7]. Flue gas from a coal-fired power plant flows through a direct contact cooler (DCC) in order to reduce the water content in the incoming flue gas and lower the temperature from 50^oC to 8^oC through a chilling water of 5^oC. Thereafter, the flue gas is sent to the first absorber in a counter-current flow of the ammonia solvent with a liquid-to-gas ratio of 4.8. An aqueous ammonia solution of 7.8 wt% is used since a higher amount could lead to the formation of solid ammonium bicarbonate in the absorption column [31, 34]. A fraction of the rich stream exiting the bottom of the first absorber is cooled and recycled back into the same absorber. This pump-around stream is fed to the first stage of the column, thereby regulating the column temperature and reducing ammonia vaporization [25, 35]. This is particularly important, as indicated by Bollinger et al. [25], as solids formation depends on the operating conditions of the absorber, based on the knowledge acquired by Alstom from other CAP experimental pilot plants.

As Li et al. [21] proposed, a single train with one-third of the total flue gas was used in the ammonia-based model simulation with the absorber diameters not exceeding 12.8 m, as recommended by both Li et al. and Chapel et al. [21, 36]. The reason for this design is to avoid uncertainties in plant construction and also for the plant capacity to be close to that of the Boundary Dam (110 MW) in Saskatchewan, Canada, the world's first commercial-scale capture plant [21, 37].

Two RadFrac absorption columns [32] (Absorber 1 and Absorber 2) were chosen based on the work of Darde et al. [35] and Jilvero et al. [26]. The CO_2 is absorbed in Absorber 1, and in Absorber 2, ammonia slip is reduced before discharge.

The flue gas from absorber 1, before entering absorber 2 as shown in Figure 3.1, is cooled down to 10^oC and fed to the top stage of absorber 2. The reason for the choice of the operating condition is to reduce clogging in the second column [38]. The gas exiting the top of absorber 1 is directly introduced at the bottom of absorber 2, allowing for temperature regulation in the column with increased loading at the top stage. The second column helps reduce ammonia slip; thereafter, the gas is sent to water wash section for further treatment. The ammonia concentration in the gas exiting the second column decreases from 12,426 ppm to 71 ppm after being passed through the water wash section before the pure gas is vented to the environment. Thereafter, the rich solution is pumped through a heat exchanger to the stripping unit where the ammoniated solution is regenerated and recycled back to the absorption unit. Part of the cold-rich solution is separated before entering the rich/lean heat exchanger through a splitter and fed to the top stage of the stripping unit; this rich-lean split technique is a proven way of decreasing the thermal energy in the regeneration unit [21]. The rich solution is preheated in the heat exchanger to 100-150°C [23] before being fed to the stripping unit. The regeneration unit operates 8 bar higher than the operating pressure of the amine process. The elevated pressure not only helps reduce the water content and ammonia slip but also helps decrease the energy penalty substantially during CO_2 compression [23].

Table 3.1:Flue gas flow rate and composition from a 500 MW coal power plant [33].

Parameter	Unit	Value
Plant capacity	MW	500
Flow rate	kg/hr	2,479,930
Temperature	°C	50
Pressure	Bar	1.016
Composition		
CO_2	Vol. %	12.33
H_2O	Vol. %	9.41
O_2	Vol. %	4.77
N_2	Vol. %	73.49

An equilibrium-based simulation method is assumed in this work. The Murphree efficiency for carbon dioxide in the absorbers is 0.1 for the ammonia- based process [35, 38].

The compression unit was modelled in four stages for the ammonia-based capture process. The inlet temperature and pressure of the process were 30°C and 13 bar, respectively. Three intercoolers were used in the compression process; they helped decrease the NH_3 concentration and reduce water content in the CO_2 product exit stream [21].

3.2.1.1 The CAP simulation

The chilled ammonia process (CAP) was simulated in this study using the electrolyte non-random two liquid (e-NRTL) method described in previous studies. The Redlich-Kwong equation of state was applied in the base method in Aspen Plus to accurately describe the vapor and liquid phases and represent their NH_3-CO_2-H_2O system behavior. An equilibrium model was used in this study for two absorbers and a stripper with a RadFrac unit operation, a process described in detail by Darde et al. [35]. The equilibrium model generally assumes complete mixing and equilibrium stages in the columns. Yul et al. [39] reported the possibility of over predicting the CO_2 absorption rate; therefore, to obtain results that resemble actual plants, we assumed Murphree efficiencies for the absorbers [35, 38]. Figure 3.2 shows the absorber profile for the mole fraction of CO_2 in the gas phase versus the liquid phase obtained in this study; this profile is a typical equilibrium profile of the absorption column [35].

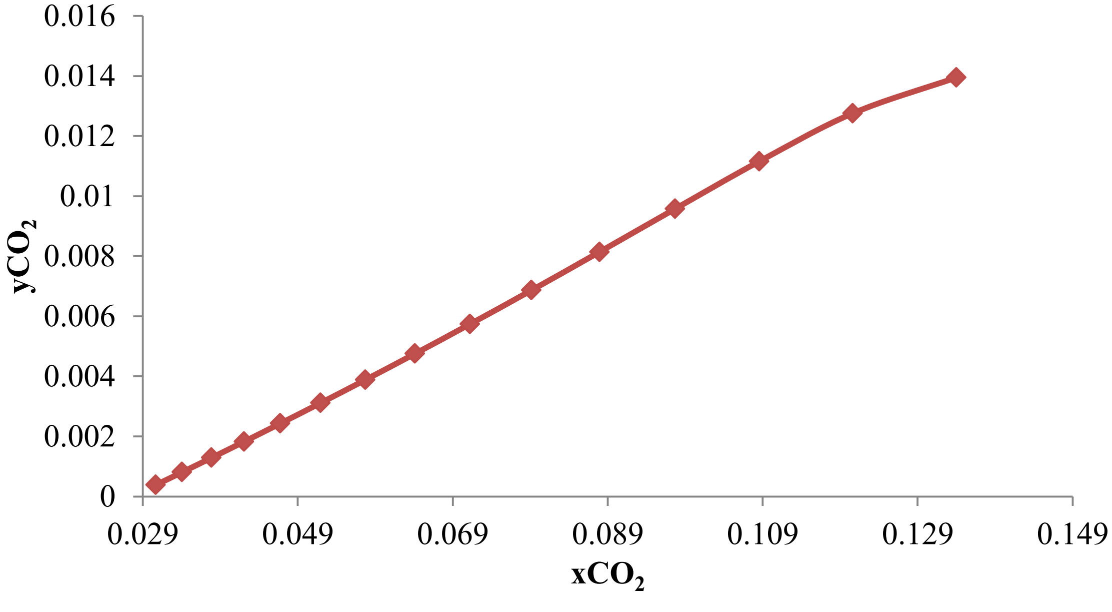

Figure 3.2: Absorber equilibrium profile in the gas and liquid phases obtained in the model

3.2.2 CO_2 capture unit

Flue gas from the power plant is assumed to be free from impurities such as SO_X and NO_X after passing through the flue gas desulfurization (FGD) unit. Staged columns were used in the absorption units (absorbers 1 and 2) to take advantage of the fast carbamate reaction and reduced ammonia volatility [26]. Furthermore, because of the volatile nature of ammonia, a separate column was modelled for the washing section that would reduce the ammonia slip before the pure gas is discharged to the environment.

The two absorbers (connected in a series) have different operating conditions since they serve different goals and purposes. The objective of absorber 1 is to absorb the CO_2 from the flue gas with the ammonia solvent while the main goal of absorber 2 is to reduce the ammonia concentration before the pure gas is sent to the washing section.

Figure 3.1 shows the schematic diagram of an ammonia-based CO_2 production with a four-stage compression unit. Absorber 1 was modeled as a 15 equilibrium-stage RadFrac column with the ammonia solvent entering stage 2 of the column. The operating temperature desired for CO_2 absorption by the ammoniated solution is 0-20^{o}C but preferably 0-10^{o}C, as recommended in Gal's patent [10]. Therefore, we assumed a temperature of 8^{o}C with a low concentration of ammonia solvent to avoid solid formation in the column. McLarnon and Duncan [40] provided insight on an ammonia-based CO_2 capture process, ECO_2, that does not show solids formation in the absorber. Though the amount of aqueous ammonia concentration used was not given, it was commonly assumed that the concentration should be between 5 and 15% [6]. Part of the rich solvent from absorber 1 was recycled back to the column via refrigeration (chilled to 5 ^{o}C); this recycling process helps regulate the target temperature to around 10 ^{o}C [17]. Most of the CO_2 is absorbed in absorber 1.

Table 3.2: Process design parameters for ammonia-based processes

Specifications	Unit	NH_3	Remarks/assumptions
Rich loading	mol/mol	0.85	[41] Range: 0.5-1
Lean loading	mol/mol	0.44	[41] Range:0.25-0.67
Reboiler temperature	^oC	144	[21]
Stripper condenser temperature	^oC	124	Obtained from model
Temperature of flue gas entering absorber 1	^oC	8	[10] Preferably 0-10 ^0C
Temperature of flue gas entering absorber 2	^oC	10	[10] Preferably 0-10 ^0C
L/G[9] ratio		4.8	Obtained from model
Desorption temperature	^oC	140	[23] [10]
Desorption pressure	bar	8	[23]
Absorber 1 pressure	bar	1.01	[34]
Absorber 2 pressure	bar	1.1	Assumed
Intercooler temperature	^oC	25	Assumed

Table 3.2 shows the general design parameters for the ammonia-based capture process. The rich solvent, with a loading of 0.85 mol CO_2/mol NH_3, is cooled to 10-15^oC before being fed to the top stage of absorber 2. Similarly, the exit gas from absorber 1 is injected in the bottom stage of absorber 2. The design parameters (see Table 3.3) of the two absorbers are not quite the same, since they have different purposes, as earlier mentioned. The second column was operated at 10-15^oC, which allows a moderate temperature to be maintained at the top stage, thereby reducing the ammonia slip [38, 42].

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[9] Liquid/gas ratio

Table 3.3: Key design parameters of the absorbers and stripper

	Absorber 1	Absorber 2	Stripper	References/comments
Absorber height (m)	15	10	11	Estimated value similar to [10] [21, 35]
Absorber diameter (m)	12	9.8	4.5	[21, 43]
Number of stages	15	10	13	Typical value[10]
Packing type	M. 250Y	M. 250Y	CMR #2	[21, 43]
Surface area (m^2/m^3)	256	256	157	[32]
Void fraction	0.987	0.987	0.98	[32]
Pressure (bar)	1.01	1.1	8	Typical value[10]

Table 3.3 lists the design parameters for the absorber and stripping column. We chose a packed column over a tray column because the former is suited for corrosive fluids; it has superior performance and better options for counter-current flows of liquid and gas for amine absorption [44-46]. The solvent exiting the absorber column is pumped to the stripping section where ammonia is stripped and regenerated, then recycled back to the capture process.

Table 3.4 gives the design parameters for the washing section for the ammonia-based capture process. Ammonia reduction in the water wash section was modeled with a 10-stage RadFrac column. The exit gas from the second absorber contains a significant amount of ammonia that needs to be reduced before the gas is vented to the atmosphere. The ammoniated solution in the gas phase was cleaned with water to obtain 71.38 ppm of ammonia concentration before discharge. Although there was slightly less exit ammonia volatility from absorber 2 than

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[10] Typical value for absorber and stripper similar to various studies

absorber 1, there was considerably less ammonia slip after the washing section compared to what was measured in the exit stream of both absorbers, as shown:

- Absorber 1: 18,217.8 ppm
- Absorber 2: 12,425.5 ppm and
- After washing section: 71.38 ppm

Various studies show ammonia slip to be from 10 to 300 ppm before discharge [17, 22, 31, 35]. The washing section achieved the aim of reducing the ammonia slip entering the washing section from absorber 2 by 99%.

Table 3.4: Washing section design parameters

	Units	Value	Remarks/comments
Number of stages		10	[35]
Condenser pressure	bar	1.5	Estimated
Column pressure drop	bar	0.03	[47]
Condenser temperature	^{o}C	40	Estimated

As shown in Figure 3.1, the rich solution exiting absorber 2 is pumped through a rich-split via a lean-rich heat exchanger to the stripper unit. The rich-split unit helps reduce the reboiler duty by splitting the cold-rich solvent through the recovery of the steam in the stripper column [21, 24, 31]. Another benefit of this modification is that the cold-rich solvent fed to the top stage condenses the water vapor exiting the stripper, thereby reducing the condenser cooling duty with a corresponding decrease in reboiler heat duty [48]. As indicated by Li et al. [21], a higher split fraction cools the stripper, resulting in a higher reboiler duty since more sensible heat is needed to reheat the split solution. Therefore, in this study, a split fraction of 0.08 was applied and led to regeneration energy of 2.46 GJ/tonne CO_2.

The stripper was modelled in Aspen Plus [32] assuming 12 stages and a kettle reboiler. The cold-rich stream is fed to stage 1 of the column while the heated rich stream is routed to the second stage at an elevated temperature of 140°C. The stripper unit operates at 8 bar with a 0.03 bar pressure drop. The cold-rich stream helps cool the temperature of the hot gas vapor and at the same time recover the ammonia from the gas phase [7]. As mentioned earlier, the rich-split process helps lower the reboiler duty and avoid solids formation in the stripper condenser [7]. A flash unit (condenser) at 30°C was used to cool the exit gas from the regeneration unit before the gas was fed to the 4-stage compression unit. The flash unit helps the CO_2 produced reach the high purity (>99%).

3.2.3 CO_2 compression unit

The CO_2 compression unit was simulated with a 4-stage compressor with inter-stage cooling. The inter-stage cooling process helps improve compression efficiency while operating in a way similar to the isothermal process [49]. Table 3.5 shows data input to the compression unit for ammonia-based capture process, with polytropic efficiencies of 84%, 80%, 76%, and 72%, for stages 1-4, respectively. The compression process is one of the most energy-intensive stages during the capture process [49, 50]. The power in the compression unit increases the thermal power plant's own needs, thereby decreasing net electricity generation efficiency [49]. It is worth noting that higher moisture content at the inlet compression stage of the capture process makes it more susceptible to degradation. Consequently, it is imperative to reduce the moisture content to the minimal as the moisture causes corrosion in the pipelines during CO_2 transportation [46].

Table 3.5: Key operating conditions for a 4-stage compression unit and compressor inlet parameters

	Unit	Value	Remarks/comments
Total flow rate at the compressor inlet	kg/hr	153,213	Model output value[11]
Mass flow rate of CO_2 at the compressor inlet	kg/hr	152,955	Model output value
Inlet temperature	^oC	30	Model output value
Inlet pressure	bar	13	Model output value
Moisture content	%	0.12	Model output value
Density	kg/m^3	24.19	Model output value
Intercooling temperature	^oC	25	Estimated
Number of compression stages		4	[51]
Discharge pressure	bar	90	
Polytropic efficiency in stages 1-4	%	84, 80, 76, 72	Assumed

The flue gas exiting the stripper unit passes through a condenser before being fed to the compression stages. The CO_2 stream temperature increases after each compression stage; therefore, the CO_2 is cooled both to decrease the energy requirement and to dry the CO_2, thereby preventing CO_2 pipeline corrosion [51]. In this study, the inter-stage cooling temperatures are assumed to be 25 ^oC and the permissible pressure drop is calculated using Eqn. 1:

$$\Delta P = \frac{(14.504 * PCO_2)^{0.7}}{10 * 14.504} bar \tag{1}$$

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[11] Output values from capture process which serves as input values to 4-stage compression unit

where ΔP is the permissible pressure drop and PCO_2 is the pressure of the CO_2.

About 59% of the water is removed following compression with a purity greater than 99% and an exit pressure of 90 bar. The pressure of the final CO_2 stream is increased to about 120 bar with a pump for transportation purposes and for the CO_2 to be suitable for further use such as enhanced oil recovery and various commercial products [49, 51, 52].

3.3 Techno-economic modelling

The results obtained from the simulated base model with corresponding mass and energy balance are loaded into the Aspen Plus Economic Analyzer (APEA) [53] for a techno-economic assessment. The plant equipment is sized and mapped in the APEA and equipment costs are estimated. The plant is assumed to be located in Alberta (a western province of Canada); the unit costs for utility, labor, and supervision are Alberta-specific rates. The total purchased equipment cost (TPEC) was evaluated from the overall cost of the equipment; thereafter, the total project investment (TPI) was calculated based on factors taken from the work of Peters et al. [54], as shown in Table 3.6.

Table 3.6: General capital investment cost factor estimates for ammonia solvents [54].

Specification	Cost estimation
Total purchased equipment cost (TPEC)	100% TPEC
Total installed cost (TIC)	302% of TPEC
Indirect cost (IC)	89% of TPEC
Total direct and indirect cost (TDIC)	TIC + IC
Contingency	20% of TDIC
Fixed capital investment (FCI)	TDIC + contingency
Location factor (LF)	10% of FCI
Total project investment (TPI)	FCI + LF

The capture plant is assumed to have a life of 20 years with a 3-year design and construction period. The construction period has 20%, 35%, and 45% [55-57] allocated to the project capital cost in the first, second and third years, respectively, as shown in Table 3.7. Furthermore, the plant operates at a capacity of 70% in the first year, 80% in the second, and 85% from the third year onward [55, 56]. The cost of producing pure CO_2, known as the product value (PV), is determined through a discounted cash flow (DCF) analysis, which is developed in an spreadsheet and takes into account cost parameters such as equipment, labor, utilities, raw materials, maintenance, operating charges, plant overhead, and general and administrative costs, for a plant capacity of 500 MWe. A 10% internal rate of return (IRR) is assumed. The cost of utilities and rates for labor and supervision were calculated based on their recent market price in the province of Alberta [58, 59]. Estimates for ammonia losses and reagent costs were obtained from Ciferno et al. [15] and Li et al. [24] as shown in Table 3.7. Unless otherwise stated in this paper, all cost values are in 2016 USD.

Table 3.7: Key factors considered in the techno-economic analysis for the ammonia process

Items	Unit	Value	Reference
Plant life	Y	20	Assumed similar to recent studies [56, 60]
Cost year basis	USD	2016	
Design and construction period			[55-57]
Year 1	%	20	
Year 2	%	35	
Year 3 and onward	%	45	
Plant capacity factors			[55, 56]
Year 1	%	70	
Year 2	%	80	
Year 3 and onward	%	85	
Maintenance cost	$	3% of TPI	[60]
Operating charges	$	25% of operating labor cost	
Operating cost subtotal	$	Sum of all operating costs including raw material and utility costs	
General & Administrative cost	$	8% of operating cost subtotal	
Internal rate of return (IRR)	%	10	
Unit cost of electricity	$/kWh	0.08	[58]
Operating hours	hr/yr	8000	Assumed similar to recent study

			[61]
Plant location		Alberta	
Operator labor wage rate	\$/hr	28.02	[59]
Supervisor wage rate	\$/hr	36.01	[59]
Solvent losses estimate	Kg NH_3/tonne	0.29	[15]
Solvent reagent cost	\$/tonne	622	[24]

3.4 Results and discussion

3.4.1 Effects of operating conditions

An advantage of CAP is its lower energy demand. As indicated in Figure 3.3, the effects of stripper pressure on capture efficiency and reboiler duty were examined. An increase in stripper pressure results in a corresponding increase in reboiler heat duty. More specifically, the heat required in the stripper to desorb the CO_2 in the rich solution increases from 2.4 to 2.9 GJ/tonne CO_2 as the pressure increases from 8 to 9.5 bar. Since high stripper pressure leads to more energy consumption by the solvent pump [21], a pressure of 8 bar was chosen for this study. It is worth noting that a high stripper pressure can enhance CO_2 regeneration rate, but the energy requirement increases [62]. High operating pressure comes with some drawbacks, such as the cost of operating high pressure steam and the need to include safety gear and install high pressure pumps.

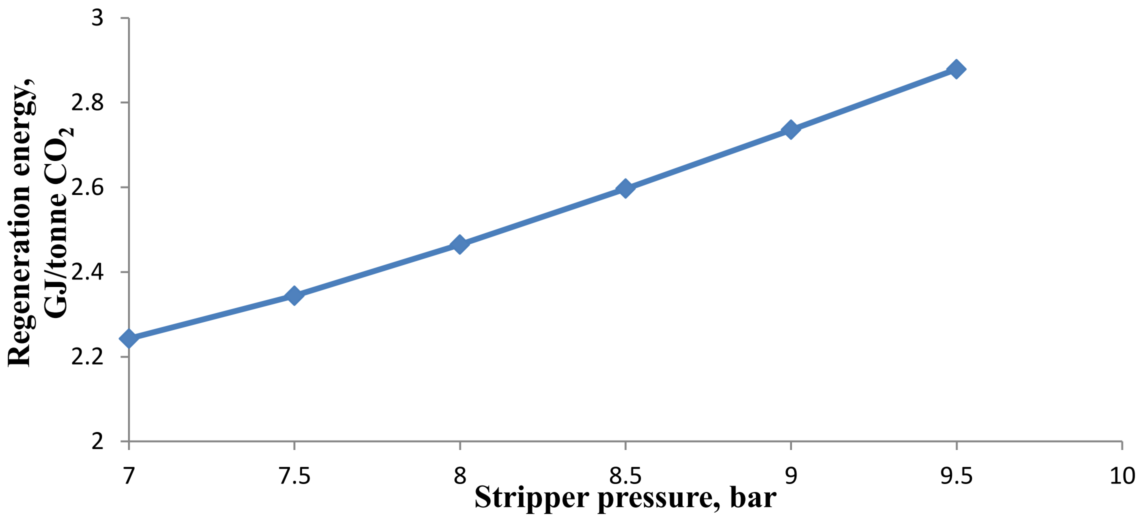

Figure 3.3: Effects of stripper pressure on regeneration energy

Ammonia slip is a great concern in the production of pure CO_2 using ammonia solvent. In order to eliminate ammonia emissions, ammonia slip needs to be reduced before the exit gas is released to the environment. The effect of a lean solution temperature on ammonia slip from the developed model is shown in Figure 3.4. An increase in lean solution temperature from 4 to 20^{o}C increases ammonia loss from 60 to 74 ppm. However, limiting the exit ammonia concentration is usually associated with an energy penalty [20]. Therefore, it is desirable to operate the CAP with a low lean temperature to control emissions and avoid solids precipitation [7]. For example, Mathias et al. [63] reported that with an absorber temperature of 10^{o}C and 26 wt% of ammonia solvent, the ammonia slip in the exit gas was 2230 ppmv. However, the ammonia slip was reduced to 242 ppm when absorber temperature was lowered to -1.1^{o}C, with 60.2% solids formation. Increasing ammonia concentration in the lean solution decreases the circulation rate due to solids formation [63]; thus, a solvent concentration of 7.8 wt% and a temperature of 9 ^{o}C were used in this research.

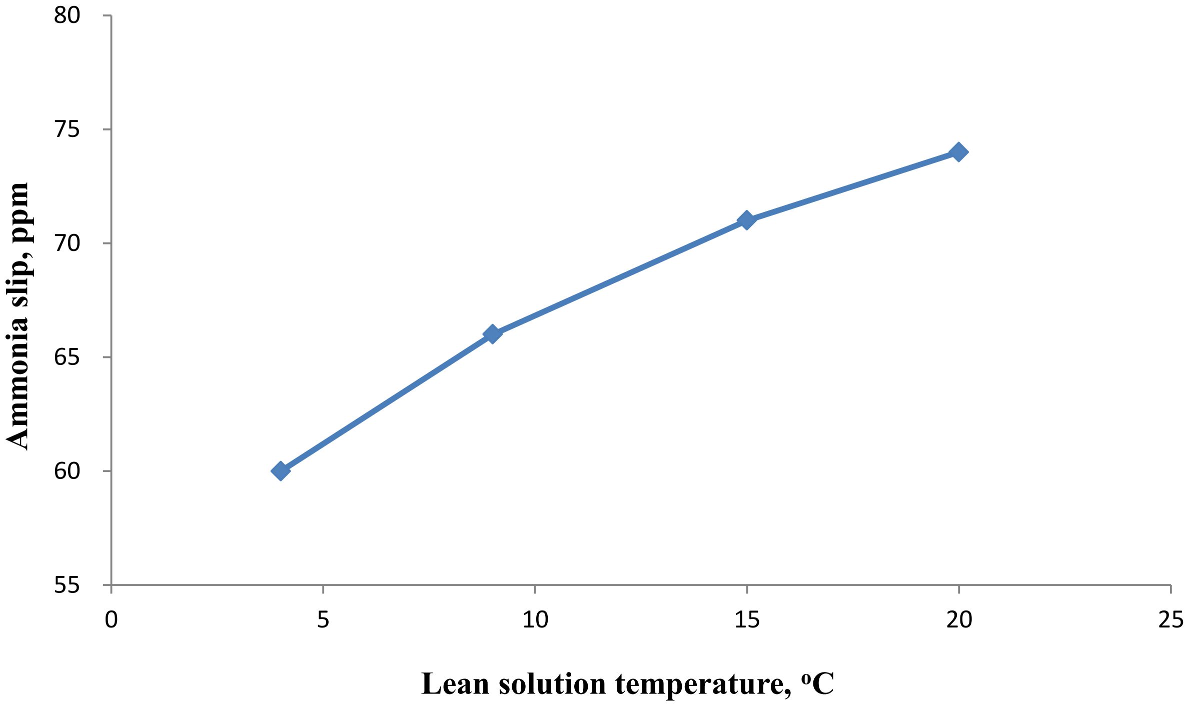

Figure 3.4: Effect of lean temperature on ammonia slip based on developed process model

3.4.2 Process analysis results

A single train simulation (167 MMe) was modeled for CO_2 production using ammonia solvent; thereafter, the simulation results were scaled to 500 MWe in an APEA model. A 90% capture rate was chosen for this study as shown in Table 3.8. The regeneration energy for the ammonia process is 2.46 GJ/tonne CO_2. The ammonia-based process is widely believed to have a lower thermal energy requirement than the MEA process; this is largely attributed to the thermodynamic properties of the NH_3-CO_2-H_2O system that allow for higher pressure during regeneration and therefore lower moisture content [6].

Since the end use of the produced CO_2 is as a feedstock for chemical conversion processes, purity greater than 99% was sought for the ammonia-based process.

A key consideration in CO_2 production is the amount of ammonia exit gas released to the atmosphere. The ammonia slip measured in this study is 71 ppm, which is in the range reported in other studies (10 to 300 ppm) [17, 20, 22, 35].

Table 3.8: Process simulation results from a 500 MWe coal power plant for 90% capture rate.

	Unit	Value
Purity	%	>99
Regeneration energy	GJ/tonne CO_2	2.46
Heat duty	MW	104.7
CO_2 exit pressure	bar	86
Moisture content	%	0.12
Ammonia slip after water wash	ppm	71.38
Ammonia slip before compression	ppm	15.13
Ammonia slip after compression	ppm	<1
Ammonia removal efficiency	%	99.4

The regeneration energy is another key output parameter in the use of ammonia solvent. The results in this study show that NH_3-based post-combustion capture regeneration energy has a significantly low thermal energy requirement as compared to MEA-based process. Darde et al. [23] for example, found that the thermal energy requirement for the ammonia process is significantly lower than for the amine process.

The degradation reaction can result in corrosion of the columns [46]. For this reason, the moisture content should be minimized in order to avoid corrosion during pipeline transportation. A 4-stage compression unit was used in both studies and the results showed that the ammonia capture process has moisture of 0.12%.

3.4.3 The cost of producing pure CO_2

The cost parameter considered in this study is the cost of producing CO_2 from a reference plant, which is the cost of CO_2 separation and compression but not transportation and storage, since the produced CO_2 is for economic uses. The plant's equipment was first mapped in the APEA model, then costs were generated, and the capital cost was calculated based on factors taken from the work of Peters et al. [54]. Figure 3.5 shows the breakdown by percentage of the equipment cost for a 500 MWe plant with a 90% capture efficiency. The equipment that contributed most to equipment cost are the absorber, compressor, stripper, and heat exchangers; these results are in line with what have been reported in other studies [5, 33, 64]. The absorber, compressor, and heat exchangers made up more than 50% of the capital requirement because of their size and energy requirement. The direct contact cooler (DCC), coolers, and chillers, grouped as "other", made up about 5% of the equipment distribution.

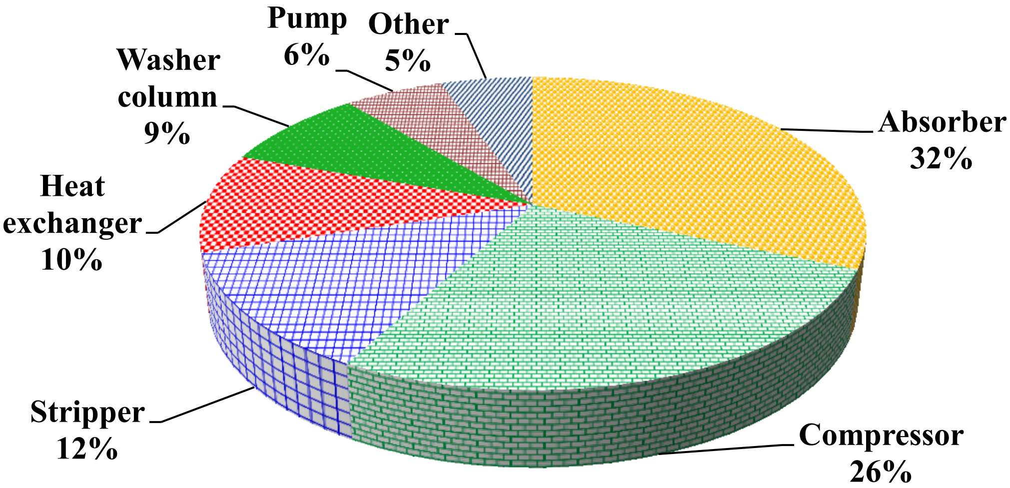

Figure 3.5: Percentage breakdown of process equipment cost distribution for ammonia-based process.

Figure 3.6 illustrates the cost breakdown of producing pure CO_2 with aqueous ammonia process. The cost for MEA process was developed earlier and has been discussed in chapter 2 of this thesis. The capital cost is 13.39 \$/tonne CO_2, which is the highest cost in the overall cost of pure CO_2 production. Table 3.9 shows the economic comparison of the ammonia-based process at a 90% capture efficiency. The raw material costs in this study for NH_3-based process is M\$ 2.88/yr.

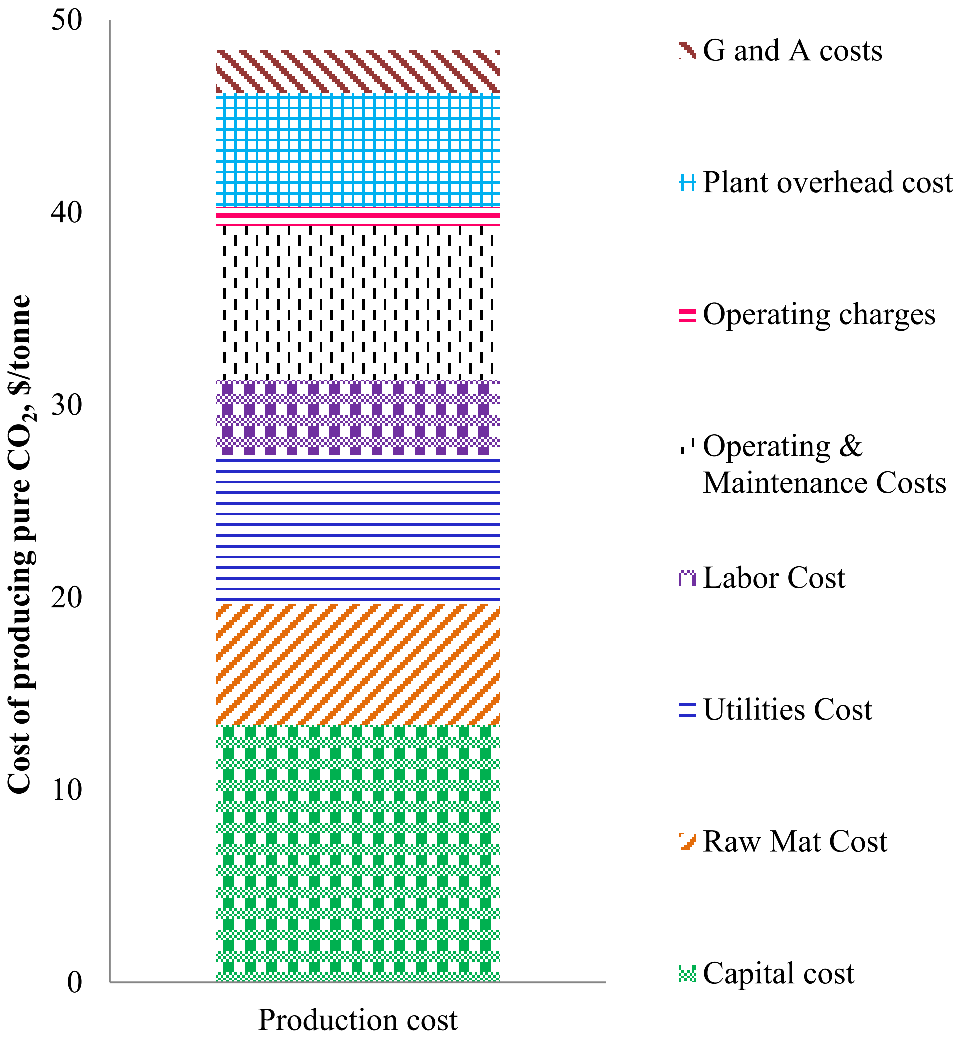

Figure 3.6: CO_2 production cost breakdown from PC flue gas for ammonia-based capture processes.

Table 3.9: Techno-economic assessment results of NH_3-based separation for a 500 MW coal power plant

Cost parameter	**Unit**	**Value**
Equipment cost	M$	150.9
Capital cost	M$	778.8
Maintenance cost	M$/yr	23.36
Labor cost	M$/yr	11.2
Utilities cost	M$/yr	22.65
Raw material cost	M$/yr	2.88
Operating charges	M$/yr	2.8
General and administrative charges	M$/yr	6.41
Operating cost	M$	34.6
Cost of CO_2 production	\$/tonne CO_2	48.42

The overall production cost from the ammonia process in this study is 48.42 \$/tonne CO_2 is comparable to the benchmark capture cost of 47.37 \$/tonne CO_2 for flue gas from coal power plant [65]. Similarly, the capture cost for a CO_2 emitting from ammonia source is reported to be 41.13 \$/tonne CO_2 [66]. These cost figures have been converted to USD 2016. The techno-economic results show that the equipment that makes up more than 50% of the capital cost are the absorption column, stripper column, compression unit, and heat exchangers same observation was reported by other researchers for capture process of CO_2 from flue gas for CO_2 avoided pathway [5, 33, 64, 67]. The cost of the two-staged absorption unit for the ammonia-based process is largely from the slow reaction rate during absorption [24], the unit sizes, and the use of two absorbers. As indicated earlier, using two columns helps reduce ammonia slip. During desorption, the increased pressure reduces the stripping cost; the elevated pressure also lowers energy economics in compression [68].

3.5 Sensitivity analysis

The effect of different parameters on the cost of producing pure CO_2 in a plant capacity of 500 MWe was examined and the results are shown in Figure 3.7. The internal rate of return (IRR) is the most sensitive parameter, followed closely by capital cost, and the least sensitive parameters are the raw material cost and operating charges. The IRR is varied from 5% to 20% [61], while the capital cost and operating and maintenance (O&M) cost is varied by 30% similar to the study carried out by Kolstad and Young [69]. However, 20% was assumed for raw material, operating charges, labor cost, plant overhead cost, General and administrative (G&A) expenses and utilities cost. The variation of capital cost by ±30% results to the product value (PV) that ranges from 41.5 \$/tonne CO_2 to 55.34 \$/tonne CO_2. The production cost can be reduced by lowering the capital cost (i.e., improving equipment efficiency and reducing equipment size), reducing regeneration energy, and increasing both CO_2 production rate and overall energy efficiency of the process.

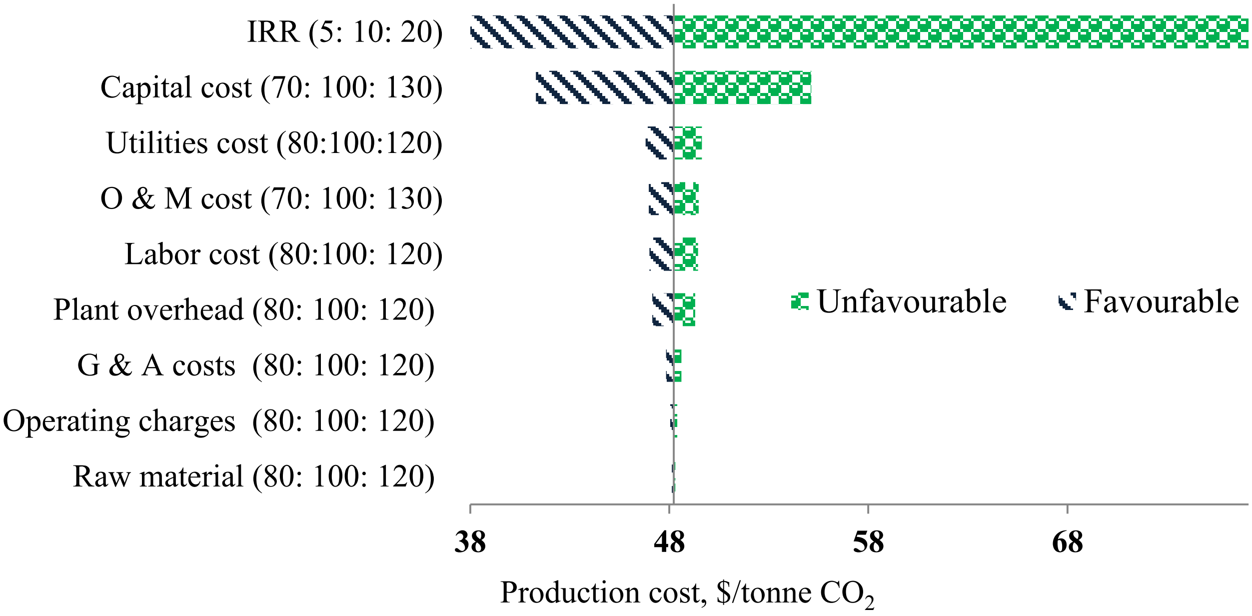

Figure 3.7: Sensitivity analyses results on CO_2 production costs in a 500 MWe capacity plant in ammonia-based process.

3.6 Uncertainty analysis

Some assumptions were made during model simulation; moreover, inherent errors and uncertainties cannot be ignored as they could affect the cost estimate. In order to assess the effects of uncertainties on the production cost, a Monte Carlo simulation was performed. The Model Risk tool [70] was run with 10,000 iterations in an Excel-based software using random values from the cost parameters that could influence production cost. Figure 3.8 shows the results of the uncertainty analysis conducted on a 500 MWe plant with a 90% capture efficiency for ammonia-based process at a 95% confidence level. The cost of producing CO_2 with ammonia solvent is 48.42 ± 3.36 \$/tonne CO_2.

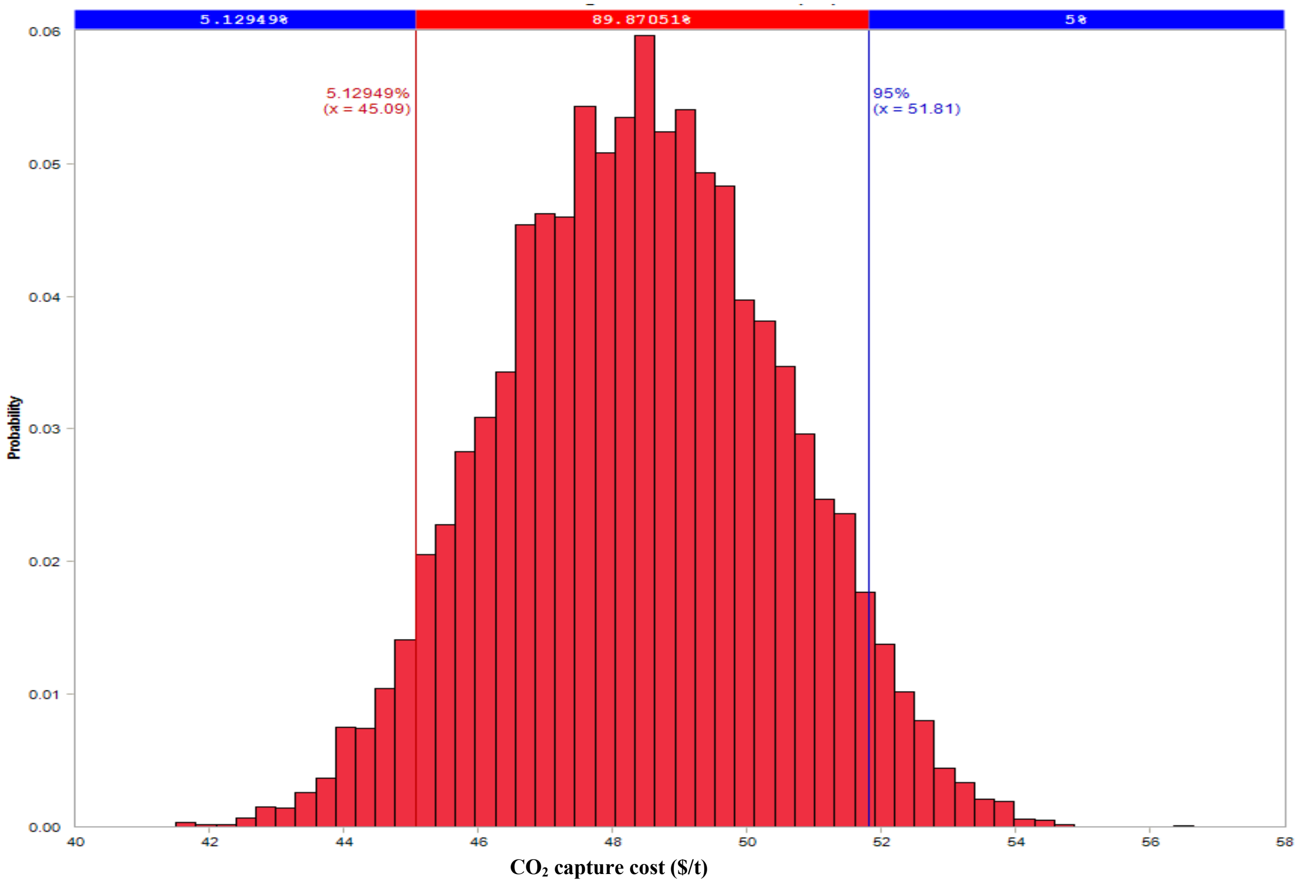

Figure 3.8: Uncertainty analyses results on the cost of producing pure CO_2 for ammonia-based process

3.7 Comparative study with published data

Most studies on carbon capture emphasize the CO_2 avoided cost; however, our focus was the cost of producing pure CO_2 from coal-fired power plant flue gases. The CO_2 production cost is usually the cost per tonne of CO_2 captured associated with the CO_2 use. In other words, CO_2 as a commodity serves as raw material to other industries. The present study estimates the cost of CO_2 production with ammonia solvent to be 48.42 ± 3.36 \$/tonne CO_2. Several studies indicate that the use of ammonia solvent result to a lower cost of CO_2 production compared to MEA solvent [7, 15, 24] as shown in Table 3.10. For example, Jilvero et al.'s [71] techno-economic analysis on carbon capture compared the use of amine and ammonia at an aluminum production plant. Their results show that the ammonia-based process at CO_2 concentrations of 7% and 10% was cost efficient, with an estimated cost of 105.72-117.16 \$/tonne CO_2, while amine technology, at low CO_2 concentrations of 3-4%, cost an estimated 132.87-138.58 \$/tonne CO_2. At the National Energy Technology Laboratory (NETL), Ciferno et al. [15] conducted a comparative economic analysis of CO_2 capture using aqueous ammonia and MEA solvent at concentrations of 7 and 30 wt %, respectively, at a 90% capture efficiency. They found the regeneration energy using MEA solvent to be 7.85 GJ/tonne CO_2 and with ammonia solvent, 3.17 GJ/tonne CO_2. In addition, the overall cost of CO_2 avoided for the MEA-based process was 61.57 \$/tonne CO_2 and 35.37 with ammonia solvent, signifying a 43% cost decrease with the use of the latter solvent. Besides having a lower capture cost, ammonia, it is widely believed, has a lower regeneration heat [24, 72-74].

Table 3.10 compares the key results of this work with three other studies [7, 15, 47]. Our results found regeneration energy of 2.46 GJ/tonne CO_2 with ammonia-based process which is in agreement with all investigations based on equilibrium-based process with heat requirement ranging from 2-3 GJ/tonne CO_2 [75]. Also, it is believed that thermal energy obtained using ammonia solvent is usually less than that of MEA-based process as indicated in Table 3.10. MEA-based process usually requires higher absorption temperature (50 ^{o}C) as compared to ammonia scrubbing (-0.15 to 20 ^{o}C); therefore requires more heat during regeneration. For this reason, the energy requirement for MEA scrubbing process should be higher than ammonia process [76]. This is similar to the results obtained in the studies listed in Table 3.10. The studies included in Table 3.10 indicate that the cost of CO_2 avoided for the MEA process is higher than for the ammonia process. The thermal energy is also higher in the MEA-based process than in the ammonia-based process. While other studies focus on the cost metrics of CO_2 avoided; the cost of producing pure CO_2 for the purpose of reuse has received little or no attention, and therefore, addressed in this study.

Table 3.10: Comparison of key results with other studies

	Ciferno et al. [15]		Yu et al. [7]		Valenti et al. [72]	This study
	MEA	NH_3	MEA	NH_3	NH_3	NH_3
CO_2 capture efficiency (%)	90	90	85	85	88.4	90
Solvent concentration (wt %)	30	7	30	6.8	20	7.8
Regeneration energy (GJ/tonne CO_2)	7.85	3.17	4	2.5	2.46	2.46
Plant capacity (MWe)	492	478	650	650	758	500
Avoided cost (\$/tonne CO_2)	61.57	35.37	101.79	66.56	53.43	48.42[12]
Capital cost (\$M)			890	800.6		778.8

3.8 Conclusion

Carbon dioxide (CO_2) emissions from fossil fuels have contributed immensely to overall greenhouse gas emissions worldwide. Carbon capture and storage (CCS) technology has received great attention over the years as a means of emissions reduction from flue gases from power plants. However, the high cost of CCS technology has slowed its commercialization considerably. Carbon capture and use (CCU) technology, therefore, is considered a means not only to reduce the carbon footprint but also to convert waste CO_2 to useful products.

This paper focuses on the cost of producing pure CO_2 from flue gases from coal-fired plants using aqueous ammonia solvents. A base process model was developed for the simulation of CO_2 production at a capture efficiency of 90% and a purity greater than 99%. The base model

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[12] Cost of production

was used for a techno-economic assessment. The ammonia-based post-combustion process was found to use 2.46 GJ/tonne CO_2. The sensitivity study found that, rate of return and capital costs have the most influence on CO_2 production costs. The costs of producing CO_2 with a plant capacity of 500 MWe found in our study are 48.42 ± 3.36 $/tonne CO_2 for aqueous ammonia. The results of this study will be of interest to investors in chemical process plants who intend to use pure CO_2 as feedstock and to decision makers across different jurisdiction including Alberta, Canada, where the carbon tax policy has been implemented, in making more informed investment decisions.

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Chapter 4: Conclusions and Recommendations for Future Work

4.1 Conclusion

The main purpose of this study was to evaluate the overall cost of producing pure CO_2 from flue gases from coal- and natural gas-fired plants with a focus on the use of MEA- and ammonia-based solvent. Given the high thermal energy demand with the use of MEA as a solvent, a comparative economic study on the cost of producing CO_2 with amine and ammonia solvents is needed. Therefore, a baseline process model was developed for the production of pure CO_2 from flue gases from natural gas combined cycle (NGCC) and pulverized coal (PC) power plants. The main sections of these plants are a Direct Contact Cooler (DCC) unit, a capture unit, and a CO_2 compression unit. Thereafter, a detailed techno-economic assessment was carried out to evaluate the cost to produce pure CO_2 with post-combustion technology using flue gases from coal and natural gas plants. Producing pure CO_2 not only mitigates GHG emissions but also provides a platform for CO_2 reuse and serves as a revenue stream to offset the capture cost in CCS technology.

The results of various capture rates (i.e., 90%, 95% and 99%) were evaluated for the production of flue gases from coal- and natural gas-fired-power plants with 30 wt% of an MEA solvent. Since the produced CO_2 in this research is intended for reuse, product purity greater than 99% was obtained from the model results across the various capture processes. For example, Enhanced Oil Recovery (EOR) processes and the food and beverage industry require a high degree of CO_2 purity. The high purity, however, requires high thermal energy. For example, regeneration energy decreased by 10% with a decrease in purity from 99% to 95%. Furthermore, the CO_2 content in NGCC flue gas is somewhat lower than PC plant flue gas and therefore requires higher regeneration energy, which affects overall production costs. As a result, the cost to produce pure CO_2 at 90% efficiency from flue gases from natural gas-fired power plants is \$79.25 ± 5.12 /tonne CO_2, which is more than \$60.61 ± 3.62 /tonne CO_2 for flue gases from a coal-fired plant. The NGCC capture process, moreover, has a higher energy intensity than a PC plant. In comparing NGCC power plant capacities of 180 MWe and 555 MWe, we found that increase in plant capacity from 180 to 555 MWe decreases the cost of producing pure CO_2 by 24%. The cost to produce CO_2 is \$104.19 /tonne CO_2 for a 180 MWe capacity and \$79.47 /tonne CO_2 for a 555 MWe capacity.

Because using MEA solvent requires high thermal energy, we compared MEA with another solvent, ammonia. Ammonia can withstand thermal degradation and requires less energy during regeneration. A data-intensive, techno-economic model was developed to compare NH_3- and MEA-based processes for the production of flue gas from a coal-fired plant. The solvent concentrations assumed in this study were 7.8 and 30 wt%, respectively, for the ammonia and MEA separation processes. The capture efficiency for both is 90% with a purity greater than 99%. The process analysis results show that the MEA-based process has a higher regeneration energy, 4.62 GJ/tonne CO_2, than the NH_3-based process of 2.46 GJ/tonne CO_2. The MEA-based process has a higher heat requirement because the solvent reaction requires a relatively high heat of reaction compared to ammonia solvent. In addition, the moisture content of the MEA process (4%) is higher than ammonia's (0.12 %), indicating that the MEA-based process is more susceptible to degradation, posing a corrosion threat during pipeline transportation of CO_2.

In the equipment cost breakdown shown in Figure 4.1 for MEA- and ammonia-based processes, we observed that the absorber, stripper, and compression unit make up more than 50% of the total capital cost, basically because of their size and thermal energy requirement. The total unit cost of producing pure CO_2 with the MEA process is 60.61 \$/tonne CO_2 and with the ammonia process is 48.42 \$/tonne CO_2. In both processes, the sensitivity study showed that capital cost and internal rate of return (IRR) are the most important cost parameters.

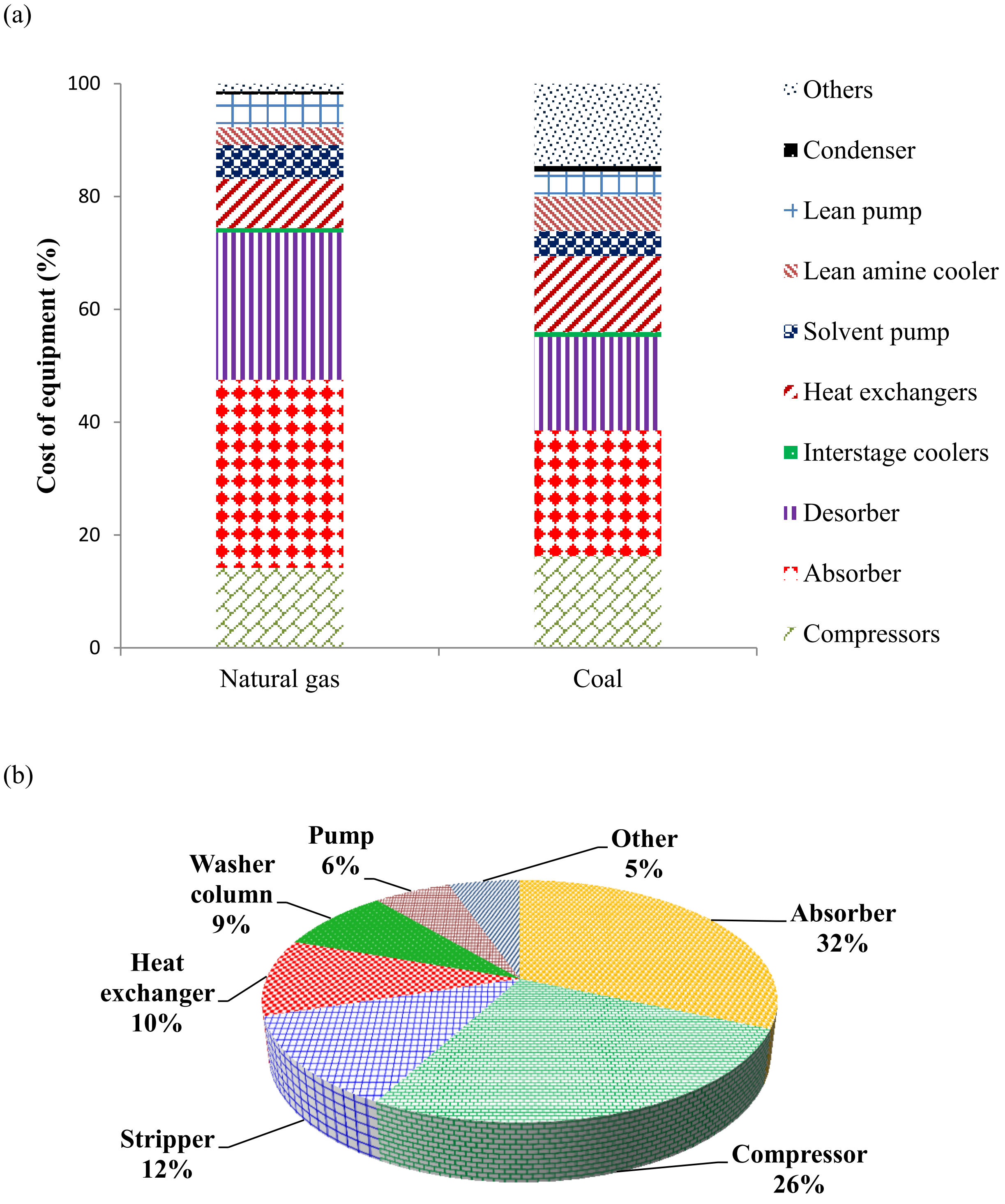

Figure 4.1: Percentage breakdown of process equipment cost distribution for (a) MEA-based process: NGCC & PC (b) ammonia-based process

The outcome of this research will be of interest not only to investors but also to decision makers in different jurisdictions, to assist in making investment decisions regarding pure CO_2 for reuse.

4.2 Recommendations for future research

The focus of this research was primarily the development of a baseline model and conduct a techno-economic assessment of the cost of producing pure CO_2 from flue gases from coal- and natural gas-fired power plants with MEA and NH_3 as solvents. Since there are few studies that assess the cost of producing pure CO_2, the author recommends the following for future consideration:

- Ammonia is a volatile chemical that is hazardous to people and the environment. During process simulation, two columns were modelled for absorption. The first column was primarily for the absorption of CO_2 gas, while the purpose of the second column was to reduce ammonia slip. The ammonia concentration exiting the second absorber before entering the washing section was a bit high due to the operating conditions. The second absorber column should be modelled to operate at a lower temperature than the temperature of the first absorber in order to reduce the ammonia slip before the vented gas is sent to the washing section.
- CO_2 production costs from emissions from other point sources such as industrial processes, waste, biomass power plant, refinery, etc., in addition to the emissions from coal- and natural gas-fired plants, should be assessed.
- This study focused on the use of MEA and NH_3 solvents in evaluating the cost of producing pure CO_2. Amine degradation and ammonia volatility are among the challenges encountered in these processes. Therefore, other separation processes such as membrane separation, ionic separation, hybrid separation systems, and others should be researched.

- A techno-economic assessment of CO_2 production together with the costs of pipeline transportation to the destination of reuse should be done; this will provide better insight for decision makers.
- The flue gas flow rate used in this study was constant. It is recommended that the rate be varied together with solvent flow rate to see the effects on thermal energy requirement, capture efficiency, and the overall cost of producing CO_2.
- Life cycle assessment should be carried out along with an evaluation of the cost of producing CO_2; this will determine if there are any environmental concerns during CO_2 production and transportation.

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