To design a process to separate the CO$_2$ from flue gases of the coal-fired boiler

UPL LIMITED

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Table of contents   **Pg no**

List of figures

List of Tables

CHAPTER-1

INTRODUCTION

1.1 NEED FOR ABSORPTION OF CO_2

An important factor in the rising global temperatures is the greenhouse gas CO_2. Since the Industrial Revolution, the concentration of CO_2 in the atmosphere has increased by 35% and continues rising gradually. If no action is done in this direction, the rate of increase in CO_2 concentration is not expected to slow down [1]. The largest of all the components of the earth's radiative forcing, according to the IPCC Assessment Report's estimation of CO_2 concentration, is a global radiative forcing of 1.66 Wm^{-2} [2]. Power plants that run on fossil fuels, particularly those that use coal as their main fuel source, are the main emitters of these emissions. The absorption and reduction of CO_2 are crucial due to its involvement in global warming. Additionally, CO_2 removal is significant and is used in many sectors today. While CO_2 is taken from natural gas to lower the cost of transportation and compression, it is necessary to remove CO_2 from hydrogen during the production of ammonia to avoid poisoning the catalyst. The removal of CO_2 has frequently been accomplished chemically using amines. However, the high flow rates of the flue gases, which include enormous amounts of CO_2, provide a significant obstacle to such systems.

1.2 OVERVIEW OF CO_2 CAPTURE SYSTEM

The CO_2 capture system used in power plants can be broadly divided into 3 categories:-
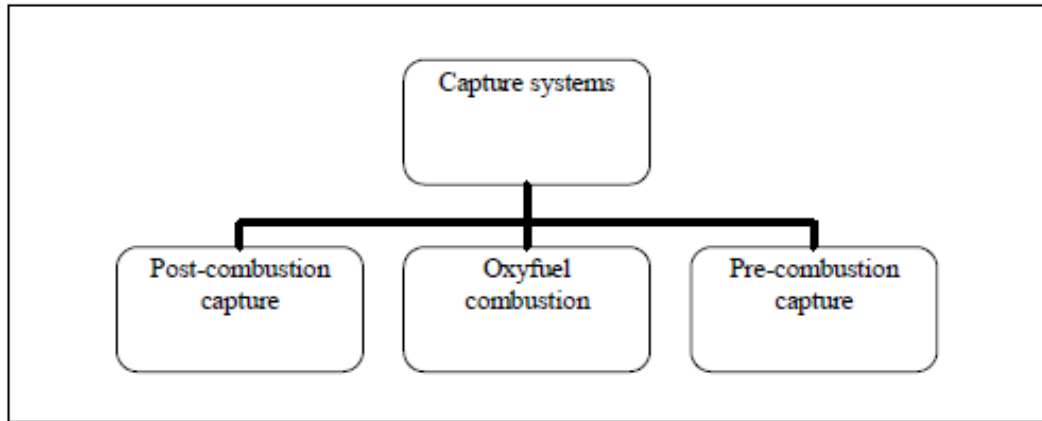
Figure 1.1: Classification of CO2 capture systems([2])

1.2.1 Post-Combustion Capture

In post-combustion capture, CO_2 is removed, once the combustion of fuel has taken place, from the resultant flue gas. A schematic of the post-combustion capture is shown in figure 1.2.
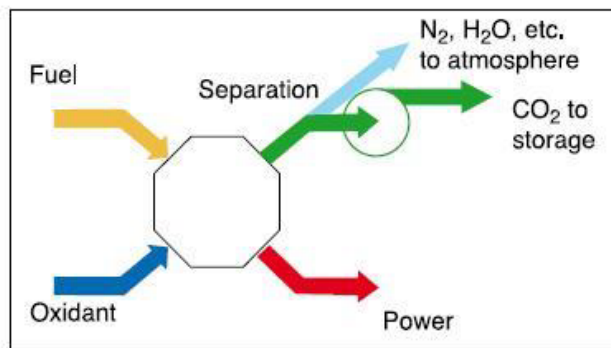
Figure 1.2: Schematic of post-combustion capture ([2])

A capture and compression system is necessary in this method. These technologies also require that the flue gas must be cleaned properly before they are fed into the capture device. The particulate matter and sulfur dioxide matter need to be removed as they result in fouling and corrosion. There are a number of methods that can be used for the post-combustion capture of CO_2 from flue gases. These include:

- Chemical absorption
- Physical absorptions

- Membrane separation
- Adsorption
- Cryogenic separation

Large-scale CO_2 removal from exhaust gases has been suggested using a variety of techniques. Cryogenic distillation, membrane separation, absorption using liquids, and adsorption using solids are the basic methods that may be utilised to separate CO_2 from other light gases. The high energy cost of cryogenic distillation prevents it from being used often to separate CO_2 from flue gases. Membranes are particularly effective for separation, especially when the components flowing across the membrane are present in high concentrations.

Typically, absorption method is employed with the use of a liquid media to separate the acid gases from the gaseous stream. In most cases, the liquid media are aqueous alkanolamine solutions or other alkaline solutions, including chilled or ambient temperature ammonia. The chemical interactions between the aqueous absorbent and the acidic gases help with absorption.

1.2.1.1 Chemical absorption
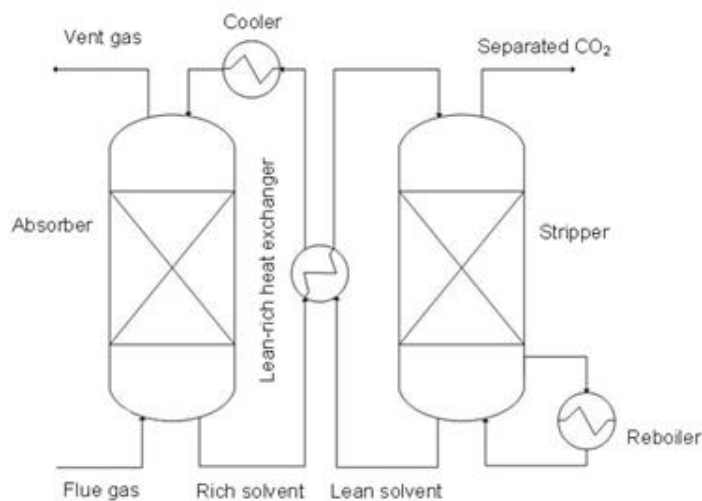
Figure. 1.3: Schematic of CO2 capture by chemical absorption

Currently, the predominant method for CO_2 post-combustion collection is chemical absorption. Aqueous alkanolamines, of which MEA is the most well-known solution, as well as alternative liquids such cooled liquid ammonia and K_2CO_3 solution can be used as the liquid medium. This mostly consists of a desorber or stripper column and an absorber. A chemical reaction involving CO_2 and an aqueous alkaline solution occurs in the absorber. This facilitates the process of

absorption. The absorbed CO_2 is removed from the rich liquid stream in the desorber with the aid of a reboiler. After being compressed to the proper pressure, the gas is transferred for storage, and the lean stream is returned to the absorber.

The heat required to produce steam, which will eventually carry away the CO_2, is provided by reboilers. In addition, energy is needed for the operation of pumps and blowers as well as for the compression and storage of CO_2. Primary, secondary, and tertiary amines are the three main subgroups of alkanolamines. Due to the low partial pressure of CO_2 in the flue gas, primary amines like MEA are regarded as the most effective and practical for flue gas cleaning among these three groups. Due to its quick reaction and lower raw material cost compared to other comparable amines, MEA is a more well-known and practical solvent at low partial pressures of CO_2.

Table 1.1: CO2 partial pressure in flue gases of different combustion systems. ([2])

Flue gas source	CO_2 concentration, % (dry)	Pressure of gas stream, MPa	CO_2 partial pressure, MPa
Natural gas fired Boilers	7-10	0.1	0.007-0.01
Gas turbines	3-4	0.1	0.003-0.004
Oil fired boilers	11-13	0.1	0.011-0.013
Coal fired boilers	12-14	0.1	0.012-0.014
IGCC after Combustion	12-14	0.1	0.012-0.014
IGCC synthesis gas after gasification	8-20	2-7	0.16-1.4 (before shift)

1.3 OBJECTIVES OF THE STUDY

- To develop a mathematical model that describes the reactive absorption of CO_2
- Optimise various parameters to get the best solvent mixture in terms of purity of CO_2 obtained, reboiler duty(energy requirements) and cost estimation

1.4 SCOPE OF THE STUDY

The scope of the study are as follows:-

- To develop a consistent flowsheet representing the actual CO2 capture and storage technique usingmixture of solvents.
- To complete the flowsheet by providing realistic input data, chemical reactions and important design specifications required for convergence and good performance.
- To study the effect of loading, capture percentage, temperature approach and other similar parameters on reboiler duty and to find important information about operational limit and other working constraints

CHAPTER-2

LITERATURE REVIEW

2.1 SOLVENTS USED FOR CO2 CAPTURE

Aqueous alkanolamines are mostly used for CO_2 absorption. As bases, these amines can interact with many acid types. They can dissolve in water because of the alcohol groups. Monoethanolamine was the first industrially utilised alkanolamine (MEA). They can be divided into primary, secondary, and tertiary varieties depending on how many organic groups are joined to the alkaline nitrogen. These diverse amines have various reaction mechanisms, products, and heats of reaction. Because the heat of absorption is higher in primary and secondary amines than in tertiary amines, they often react more quickly.

While the advantage of fast reacting amine solvent is a smaller absorber, the main difficulty is the energy required to regenerate the solvent. As a result a proper solvent must be selected. Several works are being carried out to develop mixed solvents so as to make this procedure more effective. Bishnoi (2000) [3] and Dang (2001) [4] have been working in this regard by working on Piperzine promoted MDEA and MEA respectively. Cullinane (2002) also studied the use and properties of Piperzine promoted potassium carbonate. But still more work is required and thus these mixed solvents are not yet used in industry. [5]

2.2 IDEAL SOLVENTS CHARACTERISTICS [6]

- High reactivity with respect to CO_2 – which would reduce height requirements for the absorber and/or reduce solvent circulation flow rates.
- Low regeneration cost requirements – based on a low heat of reaction with CO_2.
- High absorption capacity–which directly influences solvent circulation flow rate requirements.
- High thermal stability and reduced solvent degradation – reduced solvent waste due to thermal and chemical degradation.
- Low environmental impact.
- Low solvent costs – should be easy and cheap to produce.

2.3 SOLVENT COMPARSION

The first solvents tested in CCS came from the traditional gas treating process. The amines that have been proven to selectively absorb CO_2 from a gas stream on a large scale belong to the alkanolamine family. Triethanolamine (TEA), a tertiary amine, was the first commercial solvent that was used in the early purification gas units, followed by monoethanolamine (MEA) and diethanolamine (DEA), a primary and a secondary amine, respectively. Methyldiethanolamine (MDEA) (tertiary), diglycolamine (DGA) (primary) and diisopropanolamine (DIPA) (secondary) were also proven to be applied in CO_2 separation processes. In general, the CO_2-amine reactivity of the above-mentioned solvents shows a linear behavior in terms of the kinetics and the energy requirement for solvent regeneration, mainly depending on the chemical structure of the amine.

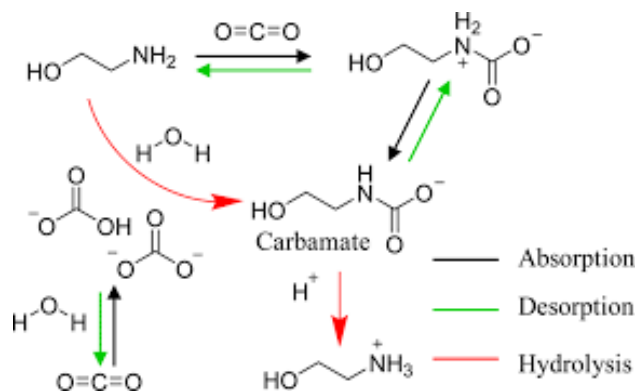
Figure. 2.1: Schematic of CO2 reaction with MEA([7])

Primary and secondary alkanolamines react rapidly with CO_2 to form carbamates.The heat of reaction involved with bicarbonate formation is lower than that of carbamate formation and thus

tertiary amines like MDEA are often blended with primary or secondary amines to reduce solvent regeneration costs. MEA is cheap; issues are degradation and high energy consumption.
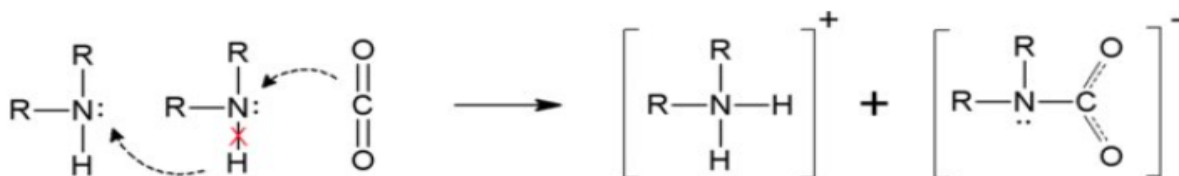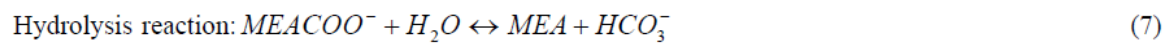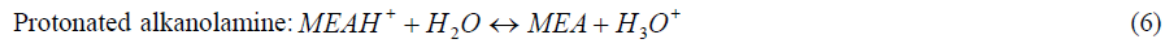

Figure. 2.2: Schematic of CO2 reaction with DEA([8])

Most primary amines have an enthalpy of CO_2 solubility about 80–90 kJ/mol CO_2, being slightly lower for secondary amines (70–75 kJ/mol CO_2). However, tertiary amines can absorb CO_2 with a significant reduction of the enthalpy of CO_2 solubility, which is usually found over in the range 40–55 kJ/mol CO_2.Primary and secondary amines provide higher CO_2 absorption rates than tertiary amines due to the carbamate formation.

Table 2.1: Promising solvents under CCS capture under investigation([9])

Solvent	Abbreviation	Type	Research institution
Piperazine/potassium carbonate	PZ + K₂CO₃	Diamine secondary	University of Texas
2-((2-aminoethyl)amino)ethanol	AEEA	Diamine primary	Norwegian University of Science and Technology
2-amino-2-ethyl-1,3-propanediol	AEPD	Diamine Primary + secondary	Korea Electric Power Research
N-methylmonoethanolamine	MMEA	Primary	Institute of Chemical Technology (India)
N-ethylmonoethanolamine	EMEA	Secondary	Institute of Chemical Technology (India) and University of Paderborn (Germany)
Piperazine/2-amino-2-methyl-1-propanol	PZ + AMP	Diamine secondary + primary (sterically hindered amine)	CSIRO Energy Tech
1,2-ethanediamine/2-amino-2-methyl-1-propanol	EDA + AMP	Diamine primary + primary (sterically hindered amine)	University of Kaiserslautern
Amino acid salts	–	–	University of Twente (TNO)
–	CASTORs	–	Institut Fraçais du Petrole (IFP)
Dimethyl-monoethanolamine/3-methylamino propylamine	DMMEA + MAPA	Tertiary + secondary	Norwegian University of Science and Technology
N,N,N,N,N-pentamethyldiethylenetriamine and diethylenetriamine	PMDETA + DETA	Tertiary + secondary	Huaqiao University
Monoethylene glycol/monoethanolamine	MEG + MEA	Primary	University of Surrey – Norwegian University of Science and Technology
Monoethylene glycol/N,N-dyetilethanolamine	MEG + DEEA	Tertiary	University of Surrey – Norwegian University of Science and Technology
Amino-functionalized ionic liquids/triethylenetetramine L-lysine	AFIL + [TETAH][Lys]	–	Huaqiao University
Carbonic anhydrase promoted potassium carbonate	CA + K₂CO₃	–	University of Kentucky
Diethylenetriamine	DETA	Secondary	North China Electric Power University
3-dimethylaminopropylamine/3-diethylaminopropylamine/3-piperidinopropylamine	DMAPA + DEAPA + 3PDPA	Tertiary	Hunan University

2.4 CHEMISTRY OF ABSORPTION

Water: $2H_2O \leftrightarrow OH^- + H_3O^+$ $\hspace{4cm}$ (1)

Hydrogen sulfide: $H_2O + H_2S \leftrightarrow HS^- + H_3O^+$ $\hspace{3cm}$ (2)

Hydrogen bisulfide: $H_2O + HS^- \leftrightarrow S^{2-} + H_3O^+$ $\hspace{3cm}$ (3)

Carbon dioxide: $CO_2 + 2H_2O \leftrightarrow HCO_3^- + H_3O^+$ $\hspace{2.5cm}$ (4)

Bicarbonate: $HCO_3^- + H_2O \leftrightarrow H_3O^+ + CO_3^{2-}$ $\hspace{2.8cm}$ (5)

Protonated alkanolamine: $MEAH^+ + H_2O \leftrightarrow MEA + H_3O^+$ $\hspace{1.8cm}$ (6)

Hydrolysis reaction: $MEACOO^- + H_2O \leftrightarrow MEA + HCO_3^-$ $\hspace{2cm}$ (7)

$$\ln K_j = A_j + \frac{B_j}{T} + C_j \ln T + D_j T$$

(8)

Table 2.2: Values of equilibrium constant equations([10])

Reaction number	A_j	B_j	C_j	D_j
Reaction 1	132.89	-13445.9	-22.47	0
Reaction 2	214.58	-12995.4	-33.55	0
Reaction 3	-9.74	-8585.47	0	0
Reaction 4	231.46	-12092.1	-36.78	0
Reaction 5	216.05	-12431.7	-35.48	0
Reaction 6	-3.038	-7008.3	0	-0.00313
Reaction 7	-0.52	-2545.53	0	0

CHAPTER-3

DEFINING THE PROBLEM STATEMENT

3.1 TECHNICAL PROBLEMS

As discussed above, thermal power plants generate a vast amount of CO_2 daily. This CO_2 when released into the atmosphere, causes global warming. Thus, companies try to use absorption-desorption techniques to remove this CO_2 from flue gas and use it elsewhere. The most popular solvent used is MonoEthanolAmine(MEA), which we have discussed in great detail.

The emerging market promises a solvent better than the conventional 30 wt% MEA. We can now use other amines like DEA and TEA. These amines require lesser energy for desorption but have less CO_2 loading and are more expensive. Thus, there is a trade-off between reboiler duty and CO_2 loading.

There are various other targets which we need to achieve simultaneously. They are summarised below:

- Max efficiency of absorber and stripper
- Least reboiler duty
- High purity of CO_2 and recycled amine
- The minimum cost of the process

These various targets that happen to be in opposite directions make this problem an optimisation problem.

3.2 DESCRIPTION OF THE PROCESS

The process can be divided into the following steps:

1. In the absorption tower, CO_2 is absorbed into the solvent (amine mixture in our case).
2. The Flue gas free of CO_2 can be safely released into the atmosphere as the purge gas.
3. The Rich amine containing the CO_2 is heated and passed through a stripper.
4. The stripper separates CO_2 from the amine and gives pure CO_2 and lean amine back.
5. This amine is then recycled and used as a solvent for the next cycle.
6. Pure CO_2 is used elsewhere.

Model Operating Parameters: *(Actual values in operation may differ)*

Flow rate: 10000 Kg/hr

Temperature: 313 K

Pressure: 1.5 bar

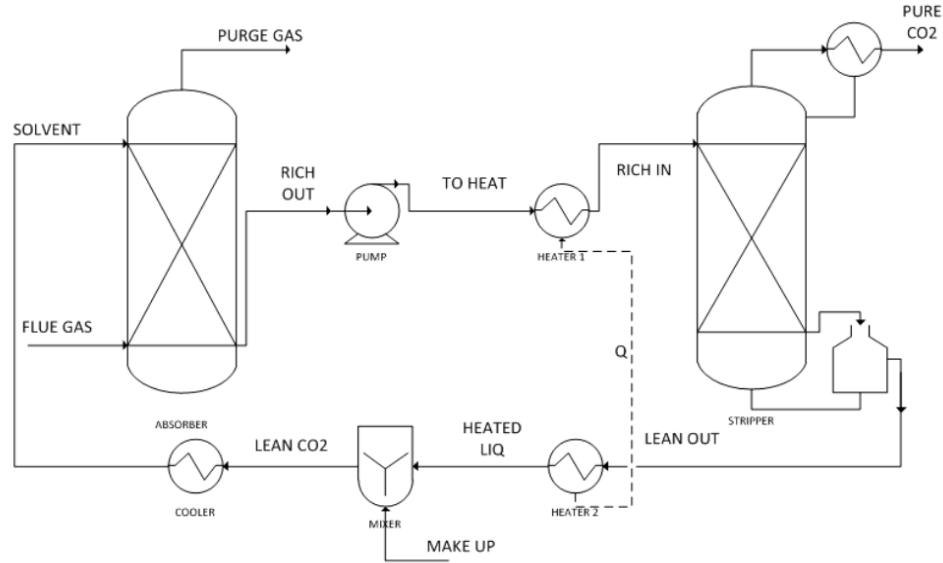A flowchart showing the Carbon capture process from the flue gas is given below:

Figure 3.1:Process flow diagram of Absorption-Stripping([10])

Table 3.1:Flue gas composition

Species	Concentration	Unit
Oxygen	6.1	mol%
Nitrogen	74.7	mol%
Water vapour	7.1	mol%
Carbon Dioxide	12.1	mol%
NOx	<5	ppm
Ammonia	<5	ppm
Sulphur Dioxide	<5	ppm
Argon	1	mol%

CHAPTER-4

MATHEMATICAL MODEL [11]

Let's take a general reaction where b_j is the concentration of component B_j

$$v_1 B_1 + v_2 B_2 \leftrightarrow v_3 B_3 + v_4 B_4 \tag{9}$$
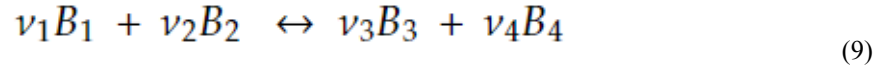
where v_j is positive if it is a reactant and negative if product

$$K = \Pi_j b_j^{-v_j}$$

For multiple reactions,

$$\sum_j v_{jk} B_j = 0$$

$$K_k = \Pi b_j^{-v_{jk}} \tag{10}$$

Assume only A dissolves from gaseous phase to liq phase

Total content of A in liquid phase α

$$\alpha = a + xm \tag{11}$$

where

x = degree of saturation

a = physically absorbed concentration

m = molarity of solution

m = equivalent concentration of species that may react equimolarly with A

Thus xm = total A absorbed chemically

Now if A (in our case ,CO_2)reacts with components B_j of the liquid phase

$$A + \sum v_j B_j = 0$$

$$\sum v_{jk} B_j = 0 \ (for \ other \ equilibrium \ reactions) \tag{12}$$

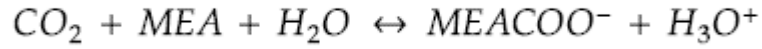both m and xm can be expressed as linear combinations of b_j

$$K = \left(\frac{\Pi\left(b_j^{-v_j}\right)}{a} \right)$$

$$\text{Also, } P_a = H*a \text{ (Henry's Law)}$$

(13)

We can then do some manipulations and get good interpretations

Our case: CO_2 absorption in MEA

$$CO_2 + MEA + H_2O \leftrightarrow MEACOO^- + H_3O^+$$

(14)

Let's say the inital molar ratio of MEA and H_2O is S (S<1)

Let initial concentration of H_2O be n_0 and MEA be Sn_0

Let concentration of final product be Sn_1

Then the final concentrations are :

$$H_2O \rightarrowtail n_0 - n_1 \approx n_0$$
$$MEA \rightarrowtail n_0 - n_1 \approx n_0$$
$$CO_2 \rightarrowtail a$$
$$MEACOO^- \rightarrowtail n_1$$
$$H_3O^+ \rightarrowtail n_1$$

The equilibrium constant is then given by

$$K = \frac{b_3 b_4}{b_1 b_2 a}$$

$$\text{Substituting, we get}$$

$$K = \frac{n_1^2}{S n_0^2 a}$$

(15)

Thus,

$$\alpha = a + n_1 = \left[\frac{n_1^2}{Sn_0^2 K} + n_1 \right]$$

(16)

MODELLING

Assumptions:

1. Only One component is removed from the gaseous phase

2. All other components are non-condensable

3. Liquid phase is non-volatile

4. Gas-Liquid equilibrium is achieved on each stage

5. The reaction between the absorbed component and the component from the liquid phase proceeds in the liquid phase.

6. No side streams enter or leave the wall.

7. No heat losses from the column.

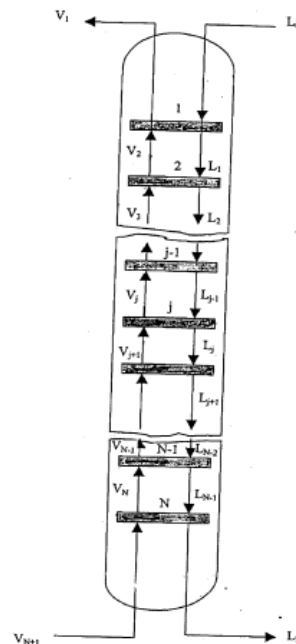8. Heat of reaction is independent of temperature.

Figure 4.1: Schematic diagram of an Abosrber([11])

Component material balance:

$$l_{j-1} - l_j + v_{j+1} - v_j = 0$$

<div align="right">(17)</div>

where l_j, v_j = component flow rate of gas leaving j^{th} stage

$$l_j = \frac{\alpha}{n_0(1+S)} L_j$$

Phase equilibrium :

$$y_j = \frac{H_{aj} x_{pj}}{P_j}$$

$$\sum_i y_{ij} = 1$$

$$\sum_i x_{ij} = 1$$

<div align="right">(18-21)</div>

Enthalpy balance:

$$V_{j+1}H_{j+1} + L_{j-1}h_{j-1} - V_jH_j - L_jh_j + \Delta H_{Rj}l_{cj} = 0$$

<div align="right">(22)</div>

where H_j = enthalpy of gaseous phase

h_j = enthalpy of liquid phase at j^{th} stage

DEGREE OF FREEDOM ANAYSIS

Equations: 5N

Unknowns : 6N ($l_j, v_j, T_j, P_j, x_j, y_j$)

DOF=N (Fix Pressure)

CHAPTER-5

FAILED ATTEMPTS

There were numerous attempts made to solve this problem and a variety of softwares were used. Some prominent methods are mentioned below. This section would be helpful to the future groups working on this problem by telling them what not to do.

1) DWSIM

   Issues and solutions tried:

   a) Can't use Standard absorption column since index out of bounds.

      i) Switched to ChemSep

   b) CO_2 absorption was too low:

      i) Need to key in the reactions. They are not inbuilt

   c) Ionic Equations can't be modeled in DWSIM:

      i) Switch to OpenModelica or MATLAB

2) OpenModelica

   Issues and solutions tried:

   a) Developed the Model given in section 4 and tested it (after multiple changes) and found division by zero error:

      i) Did Single tray analysis to avoid system of equations

   b) Still getting division by zero errors:

      i) Did manual solving to eliminate a few equations

   c) Getting unrealistic solutions

      i) Multiple roots are possible. MATLAB gave some good results

   d) MATLAB results were different for different runs:

      i) Started working on Aspen Plus

      ii) Think of it as a Design problem, not Simulation problem

3) Design problem analysis

     <u>Issues and solutions tried:</u>

    a) Was giving unphysical roots (Height of column ~ 660 m):

        i)   Did the problem again with unit corrections

    b) Data not available:

        i)   Used correlations

    c) Correlations not available for gas flows in the required range:

        i)   Completely shift to AspenPlus

4) Aspen

    a) No standard results to compare against:

        i)   Comparison with base states like no MEA or pure CO2 to be done

    b) Convergence issues

    c) Reaction mixture not following expected Chemistry

    d) Same initial conditions leading to different solutions in different trials

Thus, the future groups should try to work on AspenPlus or should try and find appropriate correlations to use the Design problem analysis described below for continuing this work.

CHAPTER-6

PROOF OF CONCEPT TESTING

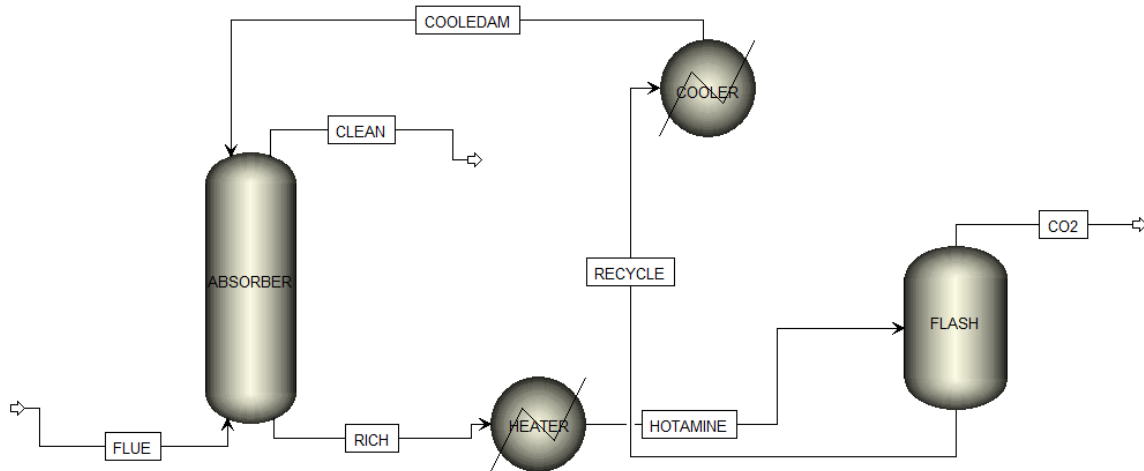Consider the flowsheet given below:

Figure 6.1: Flowsheet

The CO2 in the streams is as given below(in one of the runs):

Flue: 11126.3 Kmol/hr

Clean: 10127 KMol/hr

CO2(Stream): 2072.63 Kmol/hr

This is for a total solvent flowrate of 838.606 Kmol/hr which is 40% w/w MEA.

The operating cost for running this plant is just the Heater and Cooler Duties.

For an average Cp of 4.2 J/g K, the heat duty of a 100% efficient boiler would be:

$$= 4.2 * 2212 * 1000/3600 * (100 - 40) = 155 \ KW$$

For a 70% efficient heater, it would be 221.5 KW

A similar duty will be obtained for the cooler.

Hence, the net heat transfer is 443 KW.

Thus, at the rate of Rs 8/unit, the price of running the plant would be **Rs 3544 per hour**

The production of CO_2 is 2072.63 Kmol/hr or 91195.72 Kg/hr

The price of CO_2 from market is Rs 7/kg or **638370 Rs/hr**

Clearly using the crude analysis done above, using the CO_2 obtained from the above plant would be much more profitable than buying it from outside.

Note: Several simplifications were made in the above ballpark calculations. For example, the capital expenses are ignored and Seperation of CO_2 from water vapour isn't taken into account. Kindly consider these factors if the project is repeated.

CHAPTER-7

DESIGN ANALYSIS

OMEdit - OpenModelica Connection Editor

File   Edit   View   SSP   Simulation   Data Reconciliation   Sensitivity Optimization   Debug   Tools   Help

Writable | Model | Text View | Desin | Desin

Desin*

```
 1   model DesignProblem
 2   parameter Real D1 = 7.22e-9, D2 = 4.64e-9, C2 = 0.19*55.55*1000, C1 = 8;
 3   // Units: D1, D2 => m2/s; C2,C1 => mol/m3; y = 0.2 is assumed
 4
 5   parameter Real y0 = 0.2, yL = 0.01, G = 16, L = 20, Wg = 29, Wl = 18;
 6   // Units: G,L => lb/hr ft2   W => lb/lbmol
 7
 8   parameter Real aG = 1.97, b = 0.36, c = 0.40, Sc = 660;
 9   // Values taken for 1 inch rashchig from table 5-28 of Perry's Handbook
10
11   parameter Real al = 0.0059, muL = 2.376;
12   // Value taken for 1 inch rashchig from table 5-28 of Perry's Handbook; units: muL => lb/ft2 s
13
14   parameter Real He = 1680;
15   // unitless henry law constant
16
17
18   Real E, NTU, kya, Gm, Lm, kla, Len, HTU;
19   // E = Emhancement factor; Len = length
20
21
22   equation
23
24   E = 1 + (D2*C2)/(D1*C1);
25   NTU = 2.3*log(y0/yL);
26   Gm = G/Wg;
27   Lm = L/Wl;
28   Gm/kya = aG*(G^b)*(sqrt(Sc))/(L^c);
29   Lm/kla = al*((L/muL)^0.3)*(sqrt(Sc));
30   HTU = (Gm*(1/kya + He/(E*kla)))/3.281;
31   Len = HTU*NTU;
32
33   end DesignProblem3;
```

CHAPTER-8

FUTURE WORK

The future groups can work on one of these suggested pathways:

1. Try and get some data/reference for mass transfer coefficients of CO2 transfer to MEA

2. Try using AspenPlus in a more sophisticated system. Remember to save results after every run.

3. After AspenPlus system is established for one cycle, try using a mixture of solvents and compare heat duties

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