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OPTIMIZATION OF CO_2 REMOVAL IN AN ABSORPTION-DESORPTION UNIT

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ABSTRACT

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This review presents the optimization of monoethanolamine(MEA) regeneration in a pilot scale CO_2-MEA absorption-desorption unit. The goals of the thesis were to identify the variables affecting the stripping process and determine their optimum combination, and to give suggestions necessary for the improvement of the stripper's capacity.

A thorough perusal of the literature on the phenomena and the equipment was carried out to adequately understand the variables in action. Experiment trials were also conducted, in line with the Taguchi's method dictates, to determine the optimum combination of the tunable variables in the system.

The literature survey revealed a number of factors, four of which were further investigated to determine the optimum. The optimum condition for the washing liquid regeneration was determined. Also, two retrofitting recommendations were given to improve the capacity of the equipment.

Key words

Absorption, analysis of variance (ANOVA), design of experiment, desorption, monoethanolamine (MEA), orthogonal array, packed column, Taguchi's method.

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1 INTRODUCTION

In the past few decades, environmental consciousness has encouraged a new creed in sciences and engineering dubbed with appellations formed from their parental disciplines prefixed with green, for example, Green Chemistry and Green Engineering. This creed promotes the maintenance of greenness in the practice of sciences and engineering, that is, the integration of environmental and economical aspects of the practices. This belief is born out of the realization of the environmental impacts of the anthropogenic pollutions—pollutions resulting from human actions, chiefly from the industrial, in the past.

Global warming—a phenomenon which has been the subject of discourse in the media for years—is one of the undesired consequences attributed to anthropogenic actions. Global warming is simply the increase in the overall atmospheric temperature of the earth, leading to changes in climatic conditions. Alarms have been raised about the consequences of the earth's temperature increment; among the recent ones is the melting of the glacial ices, especially in the northern regions of the globe whose inhabitants loved dearly the winter season. By the way of explaining this warming phenomenon, hindrance to the emission of certain electromagnetic radiations caused by the concentration of the atmosphere with the so called greenhouse gases has been proposed. The inhibiting process itself is termed greenhouse effect. The greenhouse gases include carbon dioxide, CO_2, methane, ozone, chlorofluorocarbons, and water vapour. CO_2 has been regarded as a major contributor because of its large flaring from power plants and the chemical industry. In order to combat this, governments have introduced strict measures on the allowed emissions. Hence, the need for CO_2 capturing technologies has increased.

The CO_2 capturing technologies include gas absorption with solvents, gas adsorption using solids, membranes sieving, and cryogenic methods. Based on practical applications, gas absorption is far more cost effective compared to any other capturing or sequestering technology. Thus its installation dwarfs any other. Gas absorption is a famous unit operation in the chemical industry, second to only distillation, in which a liquid solvent is utilized to absorb one or more components of a gas mixture. The streams are countercurrently contacted in a packed column—a cylindrical tower equipped with solids materials in it. This unit is usually operated in a closed loop with a second tower in which the loaded solvent is regenerated and returned to the first one.

In a pilot scale CO_2-MEA absorption-desorption unit built in the Central Ostrobothnia University of Applied Sciences laboratory, the desorbing column is observed to flood frequently, a phenomenon which is known to grow the pressure drop and to reduce the desorption rate. This brings about a CO_2 build up in the whole system, significantly reducing the amount of CO_2 absorbed in the absorber. As the efficiency of the whole unit is tied to the effective liquid regeneration in the stripping tower, it is important to optimize the desorption process.

The goal of this thesis is to study the variables affecting the regeneration of the washing liquid, MEA, in the desorber of the pilot scale equipment mentioned above and then propose recommendations to help improve the capacity of the stripper; and also to determine the optimum condition of the stripping process at hand through experimentation. For the scope of this research, attention would be focused on the system at hand, that is, the study will leave out the different types of ways in which absorption can be carried out and concentrate on the way it is been done in the laboratory under study-the use of packed column. To start with, this review will begin with the discussion on the gas absorption phenomenon and

the equipment used in this study, the packed column, and then delving into the principles upholding experimental design; specifically the Taguchi's as this approach will be utilize in the study. Information on the execution, results and calculations involving the experiments will follow, and lastly, discussion and recommendations regarding the unit in question.

As regards the purpose of this work, the research questions listed below are formulated in order to further buttress the aim of the work.

1. What are the variables affecting MEA regeneration in the desorber
2. How can MEA regenerating capacity of the stripper be improved
3. What is the optimum MEA regenerating condition of the absorption-desorption unit

2 THEORETICAL BACKGROUND

Under this heading, the presentation and discussion of facts about the phenomenon of gas absorption and the use of the packed column to achieving the feat will be discussed.

2.1 Gas absorption

In chemical engineering, gas absorption (also called gas washing or scrubbing) refers to the unit operation utilized for sequestering one or more components of interest in a gas mixture. The separation is achieved by the introduction of a liquid phase, usually called washing liquid or solvent, with strong affinity for the desired component in contact with the gas mixture inside an equipment that allow for adequate interaction between the phases. Hence, the introduced liquid phase becomes loaded with the desired component(s) while the gas phase mainly contains the inert component(s). (Green & Perry 2008, 14-6; McCabe, Smith & Harriott 2005, 565; Seader & Henley 2006, 193.)

This concept has considerable applications in the chemical industry, for instance, treatment of emissions from power plants that uses fossil fuel, CO_2 and H_2S removal in natural gas processing, petroleum refinery, and coal gasification so as to avoid corrosion in the pipes and souring of the oil. CO_2 absorption makes a significant percentage of the gas absorption phenomenon because of the increase restrictions on its emission and the need for maximum usage of materials to minimize costs.

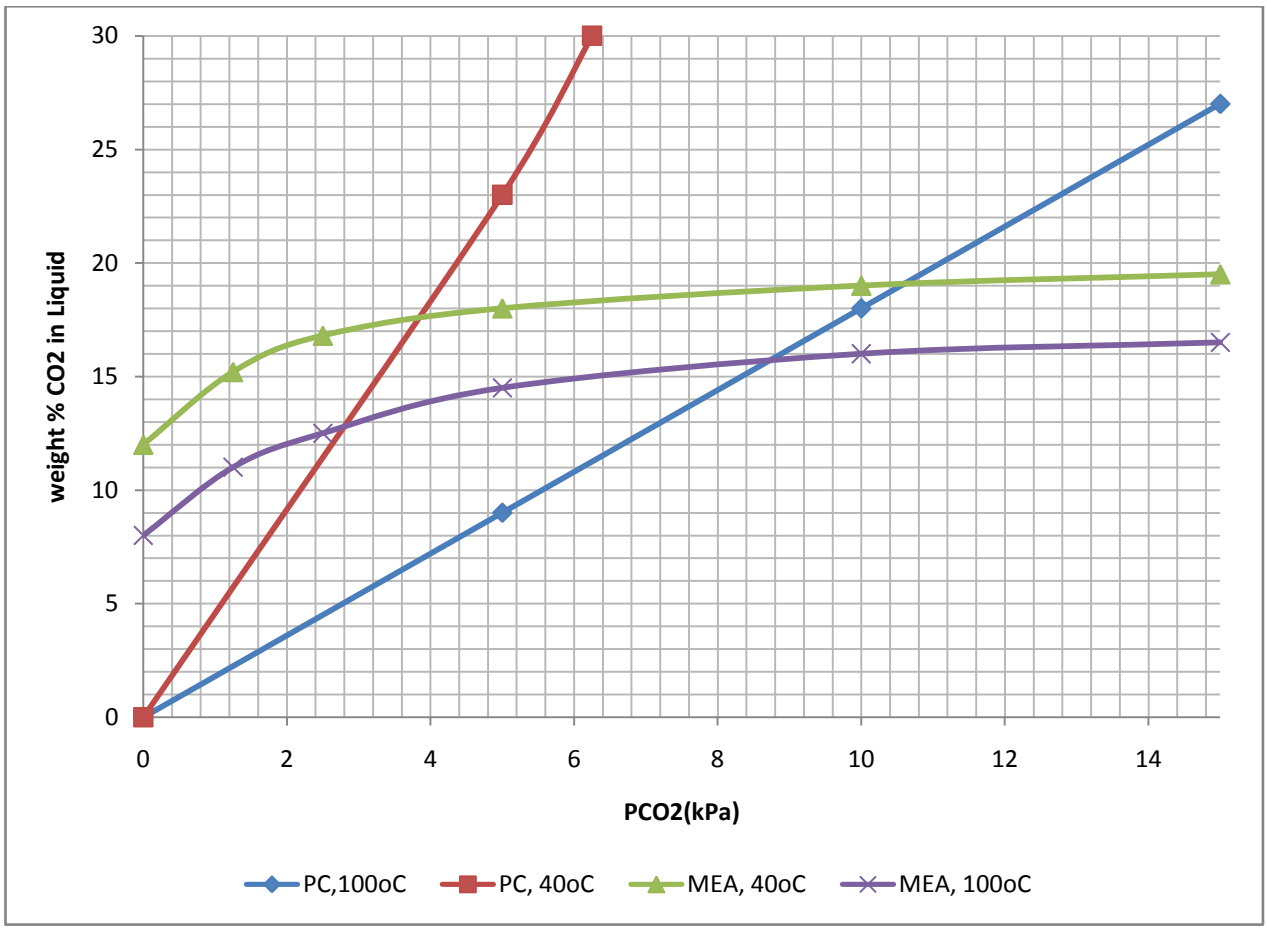
Central to the functionality of this process is adequate mass transfer, which in turn depends on proper interaction between the phases involved. With the latest development on ground, this interaction is achieved in five different ways with five different devices namely packed column, tray or plate column, bubble tower, centrifugal contactor, and the spray tower (Seader & Henley 2006, 196-200). However, it is worthy of mentioning that researchers are working on novel state of the art equipment, the fibre hollow contactor, which is supposed to be advantageous over the conventional ones though not yet commercialized (Li & Chen 2005; Koonaphapdeelert, Wu & Li 2009). The packed tower and plate column are usually the contending options to choose from when the necessary design calculations and economical considerations are critically examined side by side with installation requirements. The spray column usually finds usage when the required gas absorption process is very easy to achieve, while the bubble column is employed when a considerable residence time is required for the interaction due to either slow chemical reaction or very low solubility of the gas in the solvent. The centrifugal contactor is used when the required height of the equipment is not available or when a fast result is needed. (Seader & Henley 2006, 196-200; Zenz 1980, 289-307.)

2.2 Washing liquid

The liquid phase employed in the gas absorption process is commonly referred to as the washing liquid or solvent. In choosing the washing liquid, ''preference is given to solvents with high solubilities for the target solute and high selectivity....low volatility, low cost, low corrosive tendencies, high stability, low viscosity, low tendency to foam, and low flammability.''(Green & Perry 2008, 14-7.)

Liquid interaction with the gas phase can either be physical or chemical. Weak bonding forces between the fluids molecules are responsible for withholding the gases in physically absorbing solvent, whereas in the chemical absorption either a reversible or an irreversible reaction consumes the gases for the formation of a chemical complex. In either case, a change in the conditions of state can result in the release of the absorbed gas by the washing liquid. Chemically absorbing solvents are known to have higher solubilities compared to their counterpart as a result of the reaction taking place when the phases adequately contact each other. As the solutes disappear in the liquid phase, a chance is created for more solutes to be dissolved. (McCabe et. al. 2005, 591-604; Strelzoff 1980, 309-310.)

A comparative study of the physical and chemical absorption using the solubility data of CO_2-propylene carbonate (PC) and CO_2-monoethanolamine (MEA) under the condition of two different temperatures yields the graph below.



GRAPH 1. Linear scale plot of solubility of CO_2 in 30 weight% monoethanolamine (MEA) and propylene carbonate (PC) (adapted from Green & Perry 2008, 14-8)

Graph 1 shows a plot of the percentage by weight of CO_2 in the liquid phase (solubility) on the ordinate against the partial pressure of the CO_2 in the gas phase on the abscissa. Bearing in mind that pressure and mole have a direct relation, the plot on the x-axis is an indication of the amount of CO_2 to be absorbed. Hence, it can be concluded from the graph that physically absorbing solvents are more effective when dealing with gas mixture with high amount or partial pressure of CO_2, the reverse is true for chemical absorption. These conclusions are drawn as a

linear relationship is observed between the plots (CO_2-PC) for physical absorption, indicating a continuous increase in solubility as gas phase concentration rises. Meanwhile, the chemically absorbing plots (CO_2-MEA) exhibit higher solubility at low concentration but the solubility remains circa constant as the x-axis plot grows. (Green & Perry 2008, 14-7—14-8; Strelzoff 1980, 308-313.)

Furthermore, temperature increase is observed to decrease the solubility as seen from the isotherms in both cases. Thus, a consideration of the conditions of a gas mixture, most importantly the solute's partial pressure or amount and temperature, in addition to the good solvent qualities aforementioned, are paramount to the choice of solvent to be employed in the gas absorption unit. In addition, pressure reduction can be utilized to strip a physically absorbing washing solvent as it absorbs at a high pressure. This pressure swing application is by far cheaper than that of the temperature swing. The importance of the choice of solvent cannot be overemphasized as it is primary to the cost and greenness of the process. (Green & Perry 2008, 14-7—14-8; Strelzoff 1980, 308-313.)

2.3 Gas desorption

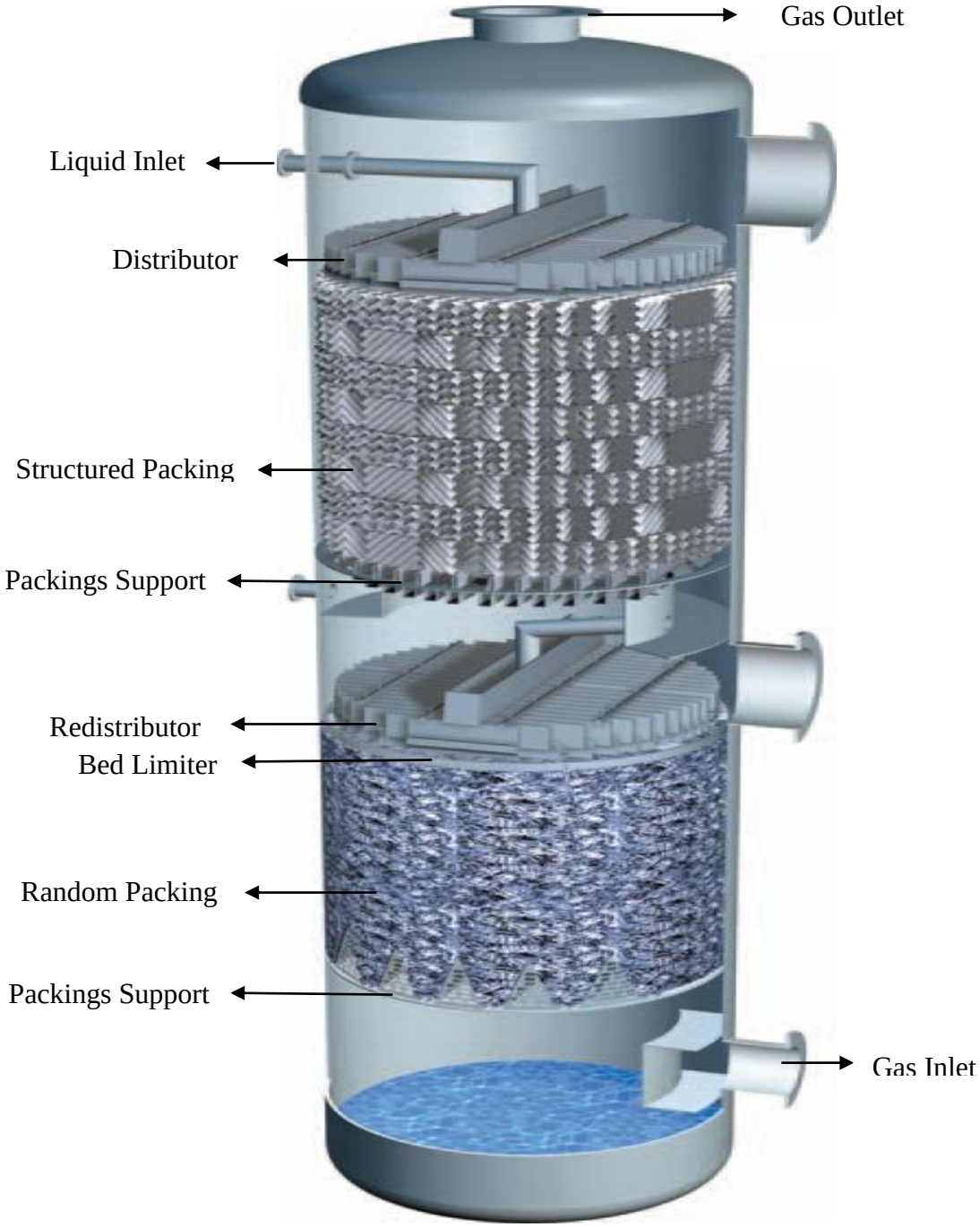
Desorption, also known as stripping, is the direct opposite of gas absorption. Here, the loaded washing liquid from the absorption column is regenerated, that is, stripped off the absorbed solute. Similarly as in the absorption process, a new phase, usually gaseous, that acts as the stripping agent has to be introduced. Depending on the nature in which the absorbed solute(s) is (are) needed; either pristine or impure, the choice of stripping agent is made to suit the desire. When the solute is to be captured, probably due to the environmental restrictions on its emission or its usefulness, steam is usually employed as the gas phase. This is

rather expensive but the choice strongly relies on the weight of the benefits or regulations on ground. On the other hand, when it is not necessary to recover the solute in a pure state, either to be flared or captured as well, a cheaper gas flow is made use of as the stripping agent. Air is commonly used because of its availability. (McCabe et. al. 2005, 590-591.)

Desorption is run in a continuous closed loop, hand in hand with the gas absorption process. The regenerated or lean solvent from the desorber is repumped into the absorber as a fresh solvent. This fact underlines the importance of effective regeneration in the desorber as its core to the overall efficiency of the unit as a whole. The packed column as well is commonly in use for the process of desorption. (Seader & Henley 2006, 193.)

2.4 The packed column

The packed column/tower is a device employed in the chemical industry for a number of mass transfer operations. Common operations carried out with the packed column include distillation, gas absorption and desorption, and leaching. From an external view, the packed column is no more than a cylindrical tower equipped with two openings at the top and bottom to allow for liquid and gas streams entry and exit. These streams are arranged countercurrently with the liquid flow's inlet and outlet situated at the top and bottom respectively. The converse is true for the gas flow. An internal reconnoiter from top to down reveals some additional internal features such as liquid distributor, section(s) of packings supported above and below with a support plate, liquid collectors, and liquid redistributors. Graph 2 shows a packed column with its internals visible. (Green & Perry 2008, 14-53; McCabe et. al. 2005, 565-566; Seader & Henley 2006, 198.)

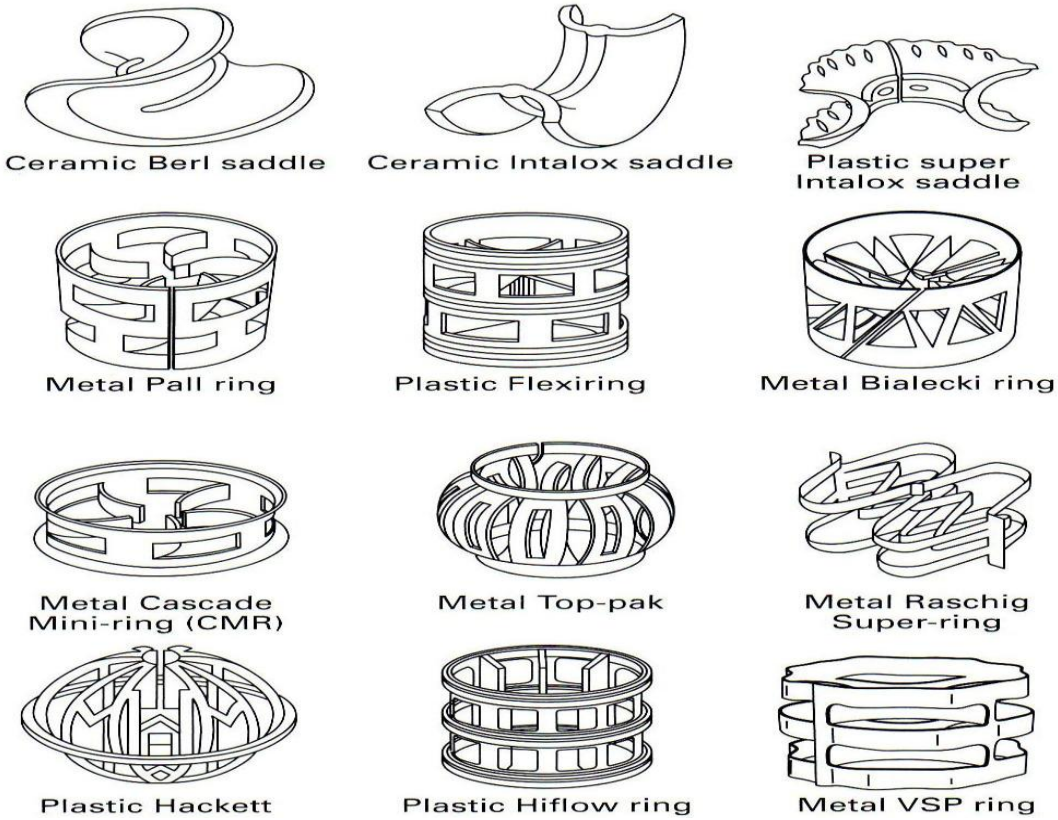


GRAPH 2. An internal view of the packed column (from Sulzer Chemtech's internals for packed column)

2.4.1 Packed column internals

The column internals are discussed in this heading. The distributor is put in place to distribute the inlet liquid flow evenly across the packing's surface. The liquid inlet feed is usually directed to the distributor, which then effects the distribution. The distributor plays a vital role in the overall efficiency of the column, especially in columns with a large diameter, as its presence is necessary to provide adequate contact of the phases. (Green & Perry 2008, 14-73; McCabe et. al. 2005, 565; Saint-Gobain Norpro 2001; Seader & Henley 2006, 198.)

Packings are solid objects gathered in sections inside the packed column. They are available in different materials and shapes from which they are usually named, for example, ceramic Raschig ring. Packings are shaped with the goal of increasing the interfacial surface area and reducing the pressure drop as fluids flow through them. The material with which packings are made of determines their wettability, corrosion resistance, strength and price. All of these are significant factors that are considered when making choice of packing. The oldest packings used are the Raschig rings and the Berl saddles. These have been largely replaced by varieties of design, in terms of structural improvement, to handle better flow capacity, to produce lower pressure drop, and to exhibit much more surface area for fluid interaction. Some of the existing packings types are shown in Graph 3 below. (Green & Perry 2008, 14-53—14-54; McCabe et. al. 2005, 565-567; Seader & Henley 2006, 198-199.)



GRAPH 3. Various packing types(Seader & Henley 2006, 199)

An inverse proportional relationship exists between packing size and pressure drop in the column, also with the mass transfer performance. However, a high mass transfer rate and low pressure drop are the required conditions in view of the performance of the column and the cost of pumping respectively. The mass transfer advantage rendered by smaller packing is said to be minimal; consequently, larger packings are favoured because of their lower pressure drop edge. The general rule of thumb in choosing packing size is that packing size should be about one-eighth of the column's diameter (McCabe et. al. 2005, 568-9; Seader & Henley 2006, 198-199). Adherence to this rule is said to eliminate channelling, the maldistribution of liquid throughput, which is a major problem in towers with a large diameter. Additionally, as liquid distribution is vital to the efficiency of mass transfer in packed columns, sections of packings allotted with distributors or re-

distributors are used especially in tall columns also to ensure even distribution. According to Saint-Gobain Norpro's packed tower internals guide (Saint-Gobain Norpro 2001), sections of packings should not be more than fifteen times the column diameter and contain less than twenty theoretical stages, while Zenz (1980) proposes the smaller one of either five times the column's diameter or ten feet packings height. (McCabe et. al. 2005, 568-9; Seader & Henley 2006, 198-199; Zenz 1980, 289-307.)

Packings can be either dumped or arranged into the columns. The former is referred to as random packing while the latter can either be stacked or structured packing. Stacked packings are arranged in the column by hand, whereas structured packings are made pre-arranged by the manufacturer. Structured packings usually give less pressure drop compared to random packings, but random packing gives higher mass transfer. This statement is consistent with the results of the experiments conducted by Longo and Gasparella (2009) on desiccant regeneration performed with both random and structured packings. Under their experimental conditions, structured packings gave a 65-75% lower pressure drop while random packing had 20-25% higher regeneration performance. Apparently, due to these facts, structured packing can handle bigger streams, but it is expensive as compared to its counterpart. This is why random packing is found to be more cost effective in the smaller towers. When dealing with very high flows, structural packings are essential for the sole reason of extending the flooding capacity and to minimize the energy spent on pumping. (Green & Perry 2008, 14-53—14-54; McCabe et. al. 2005, 568-604; Seader & Henley 2006, 198-199; Longo & Gasparella 2009.)

The packing support is the seat on which the packings gain their stance. Packing supports commonly possess a netlike surface to allow for the passage of the gas

phase upward the column. Similar in structure is the bed limiter which is placed on top of the packings to keep them orderly when gas flow is increased close to or above the fluidization value. Every section of packings has a packing support on which to stand and a limiter to maintain order above it. (Saint-Gobain Norpro 2001; Seader & Henley 2006, 198.)

In order to avoid channelling, the idea of using sections of packings was introduced. A recommendation as to the height of packings to be utilized at a section has been mentioned earlier. The collector serves to gather the downward coming liquid phase from the upper packing section and leads it to the redistributor which wets the next packing section. (Green & Perry 2008, 14-53—14-54; McCabe et. al. 2005, 568-604; Saint-Gobain Norpro 2001; Seader & Henley 2006, 198-199.)

2.4.2 Absorption-desorption in the packed column

Having explained the absorption and the desorption phenomena in the early part of this chapter, and described the equipment, packed column, which can be employed for both processes, it is imperative to talk about how the duo phenomena are united in the twin towers working together as a continuous process to forming the unit operation dubbed gas absorption. The gas absorption unit normally consists of two towers; one for absorbing the target gas, the absorber or scrubber, and the other for regenerating the used solvent, the stripper or desorber.

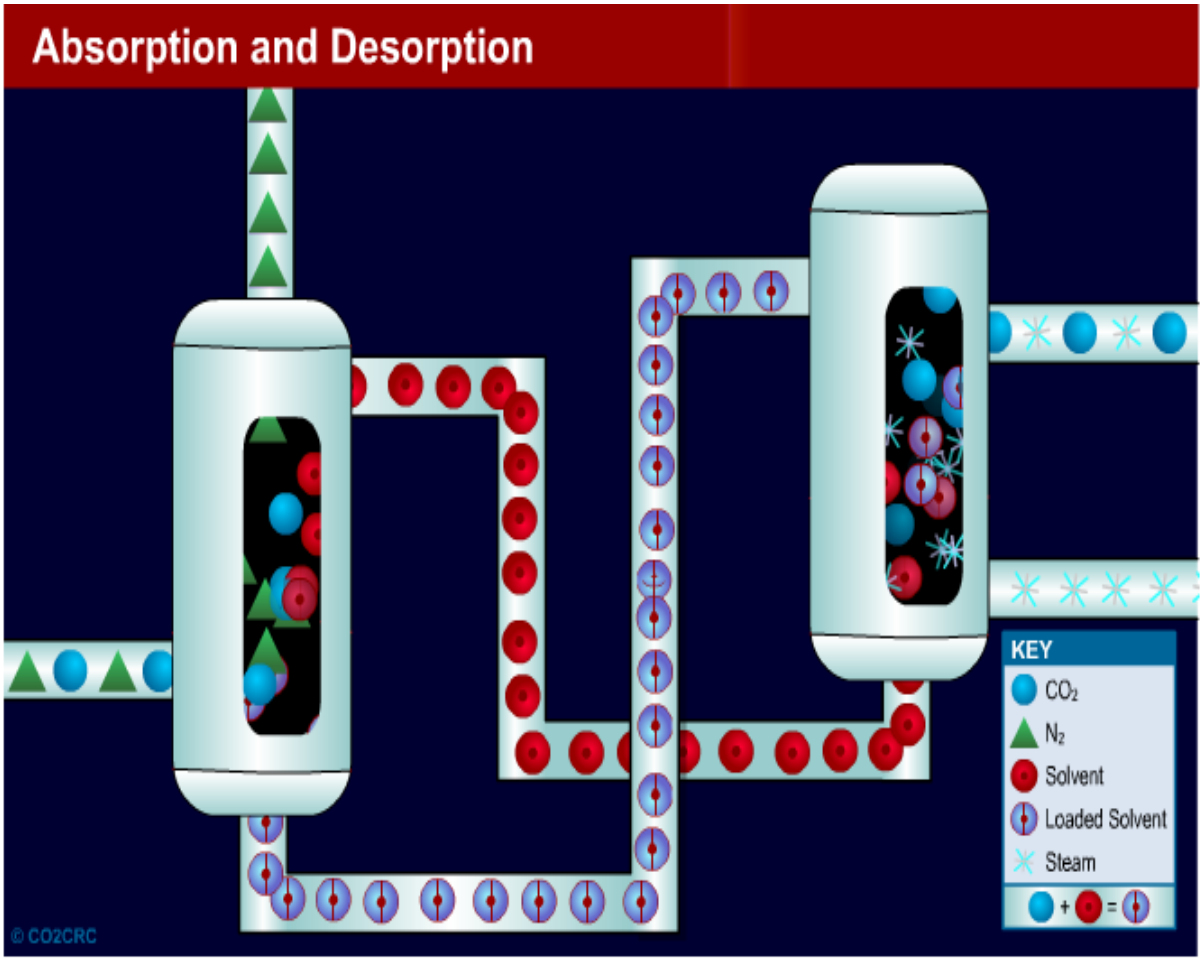
The absorber is a packed column with all the ancillaries aforementioned. On the introduction of the solvent from the top of the column, the distributor spreads it as evenly as it can over the cross sectional area of the packings directly below it. As the solvent is pulled down by gravity through the sections of packing, the

throughput is broken into droplets and spreads around the interfacial area of the packing; simultaneously, the inlet gas mixture to be purified would make its way up from the bottom of the column also through the packings (Zenz 1980, 289-307). When the fluids meet at the surface area provided by the packings, they interact and mass transfer of the target gas will take place at that instant. This happens continuously as the solvent moves through the packings. Consequently, the liquid phase increases in the amount of the absorbed gas down the column whereas the gas phase reduces in its concentration of the solute up the column. Hence, the outgoing gas is purified, or at least contains less of the target gas; on the other hand, the outgoing liquid phase is loaded with the absorbed gas. The need to regenerate and reuse the spent washing liquid usually invokes the use of a second tower, known as the desorber. (CO_2CRC 2010; McCabe et. al. 2005, 565-569; Seader & Henley 2006, 198; Zenz 1980, 289-307.)

The desorber as well is merely a packed tower except that it is equipped with a reboiler for the production of steam as the gas phase when the target gas is to be captured purely. When the target gas is not needed pure and there exist no restriction on its emission, a cheaper gas can be utilized. Just like in the absorber the liquid inlet, the loaded solvent in this case, is from the top the column while the gas phase, the stripping agent is from the bottom. In the same manner, they meet on the packings and the absorbed gas is released into the gas phase. Here, the desorbed gas is collected on its way out of the column while the fresh solvent is sent back to the absorber, making a cyclic process. (CO_2CRC 2010; McCabe et. al. 2005, 565-569; Seader & Henley 2006, 198; Zenz 1980, 289-307.)

Generally, a state of high pressure and low temperature favours the absorption process. However, high temperature and low pressure suits the desorption phenomenon. A heat exchanger is usually installed between the towers with the pur-

pose of heating up and cooling down the respective circulated solvent. Shown in Graph 4 below, is a gas absorption unit where CO_2 is absorbed from its mixture with nitrogen. A number of details were left out concentrating only on the flows and the material exchange.



GRAPH 4. An absorption–desorption unit (adapted from CO_2CRC 2010)

2.4.3 Packed column design

In this subtopic, light will be shed on major issues pertaining to the decision of sizing the packed column. Sizing of the equipment refers to how wide and high the diameter and height of the column would be respectively. Prior to the deter-

mination of these two important sizing-parameters, series of decisions would be made. Some of these are backed by sound theories whereas others can be regarded as an act which can only be acquired with years of experience.

Customarily, when the need arise to employ the packed column, say for the absorption of a contaminant from a flue gas, the following parameters are specified; the gas mixture's composition, flow rate, temperature and the exit gas composition required. The problem posed to the designer is threefold as stated below:

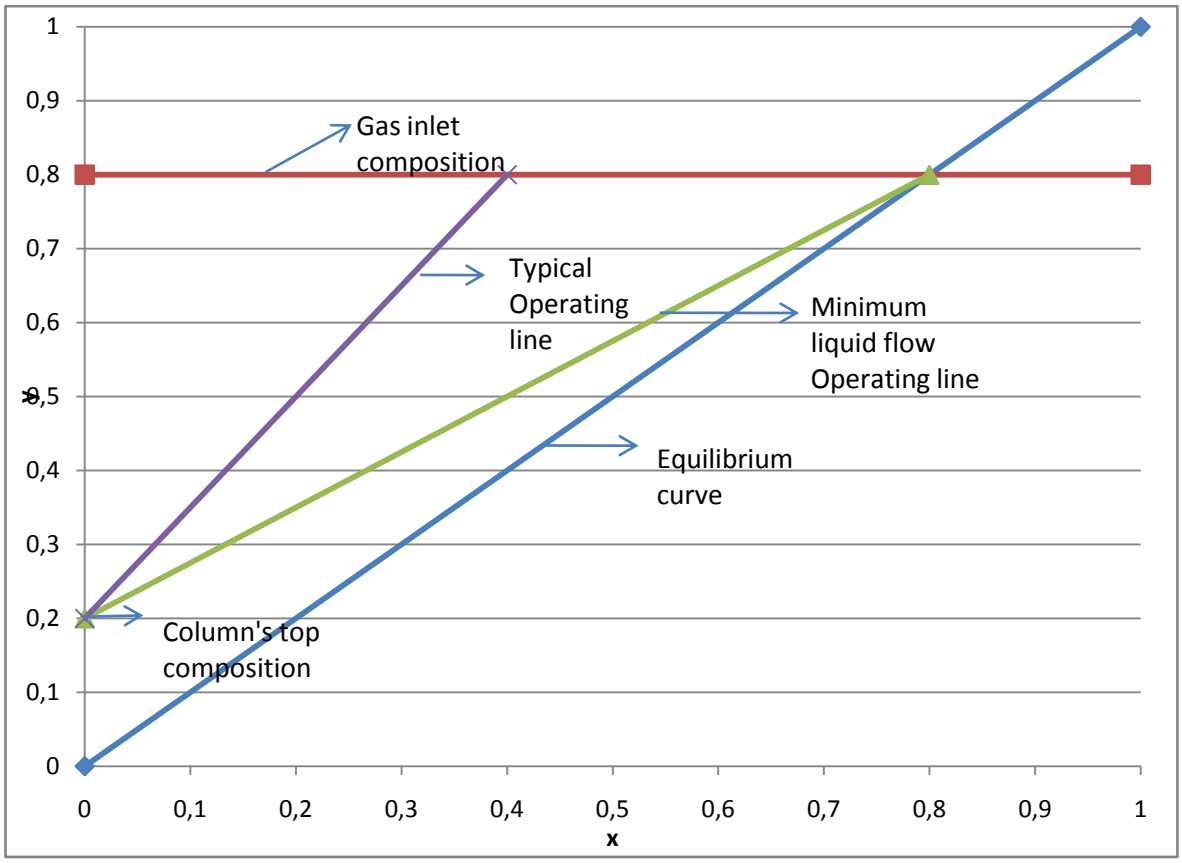
1. Choice of solvent to utilize
2. Determination of column's diameter
3. The required height of packings

The tedious process involved in deciphering the problems stated above would be treated in turns.

The choice of washing liquid is the starting point for the designer. In section 2.2, an extensive discussion was made on washing liquids. This includes the properties expected of a prospective solvent and the conditions favourable for physically and chemically absorbing solvents. Knowing fully these facts, what the designer needs to do is check his database of washing solvents in literature and look up the appropriate solvent to suit the conditions at hand.

Having made the choice of solvent, the designer ought to check the feasibility of the task before proceeding further. In order to affirm this, a reliable source has to be consulted to get the equilibrium information of the fluids at the operation temperature, pressure and concentration. Equilibrium data abound in various forms in literature. On a carefully graphed equilibrium data, the prospective composition at the top of the column is located and the gas inlet mole fraction is depicted

with a horizontal line as illustrated in Graph 5. Any line joining the located point and touching the horizontal line is referred to as the operating line.



GRAPH 5. Equilibrium and operating line diagram (adapted from Seader & Henley 2006, 202)

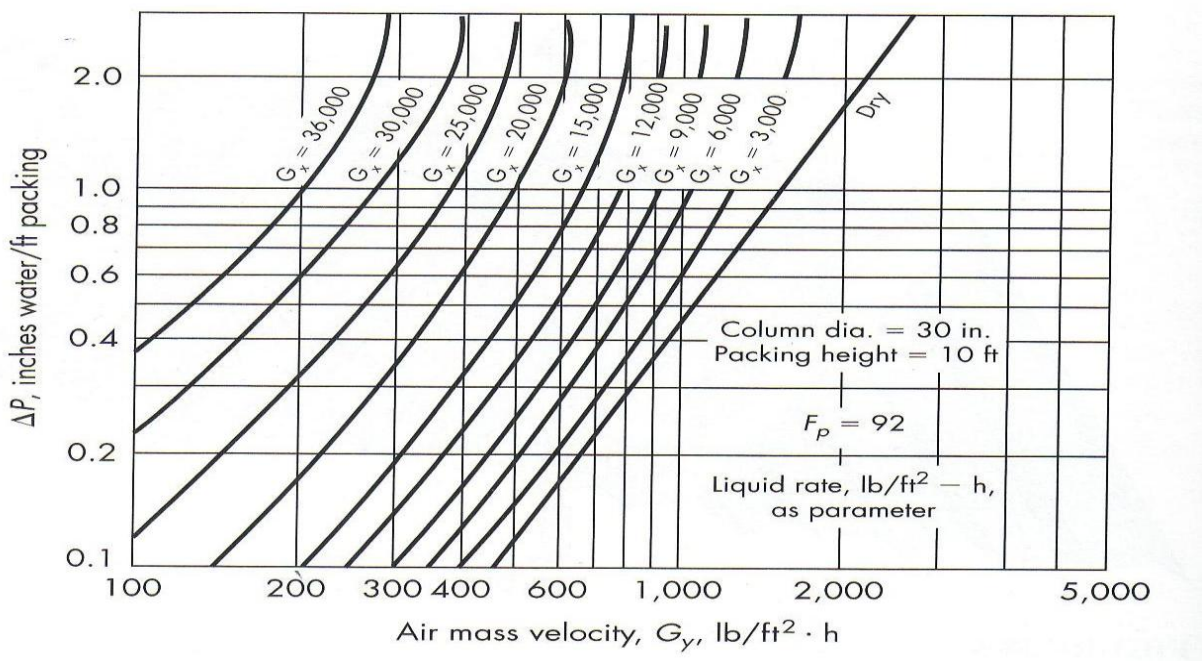
For the proposed absorption to be possible, the operating line must be above the equilibrium curve. For the stripping process, the operating line must be below the equilibrium curve and the solvent will lose the solutes until there is no more driving force- the vertical distance between the curves. Hence, the fluids will exchange material until the equilibrium condition is achieved, ideally. (Green & Perry 2008, 14-10; McCabe et. al. 2005, 576-578; Seader & Henley 2006, 201-203.)

If a green light is gotten from the feasibility test, then the next thing is to decide the forthcoming quantity of liquid throughput to pass through the column. It is necessary to mention at this point that the slope of the operating line is the ratio of the molar flow rate of the liquid phase to gas phase; more detail will be given latter about the derivation of this fact. Bearing this in mind, it is evident that a reduction in the value of liquid flow rate, when other variables are kept constant, will lean the operating line towards the equilibrium line. As far as absorption is concerned, the operation line must remain above the equilibrium line. Therefore, the lowest liquid flow attainable is achieved when the operation line just meets with the equilibrium curve as seen in Graph 5. The liquid flow at this point is called the minimum liquid rate. Likewise, a minimum value exists for the stripping agent utilized in the desorber which can be located in a similar manner but on the opposite side. At the minimum rate, an infinite number of stages are required to accomplish the necessary absorption. This throughput might not be enough for adequate wetting of the packings. The minimum rate notwithstanding, the actual flow has to be chosen to actualize good distribution in the column. The general rule of thumb is to make the actual flow a multiple of the minimum, ranging from 1.1 to 2. Multiples of 1.2 to 1.5 are said to be more cost effective, however. (Green & Perry 2008, 14-9; McCabe et. al. 2005, 578; Seader & Henley 2006, 202-203.)

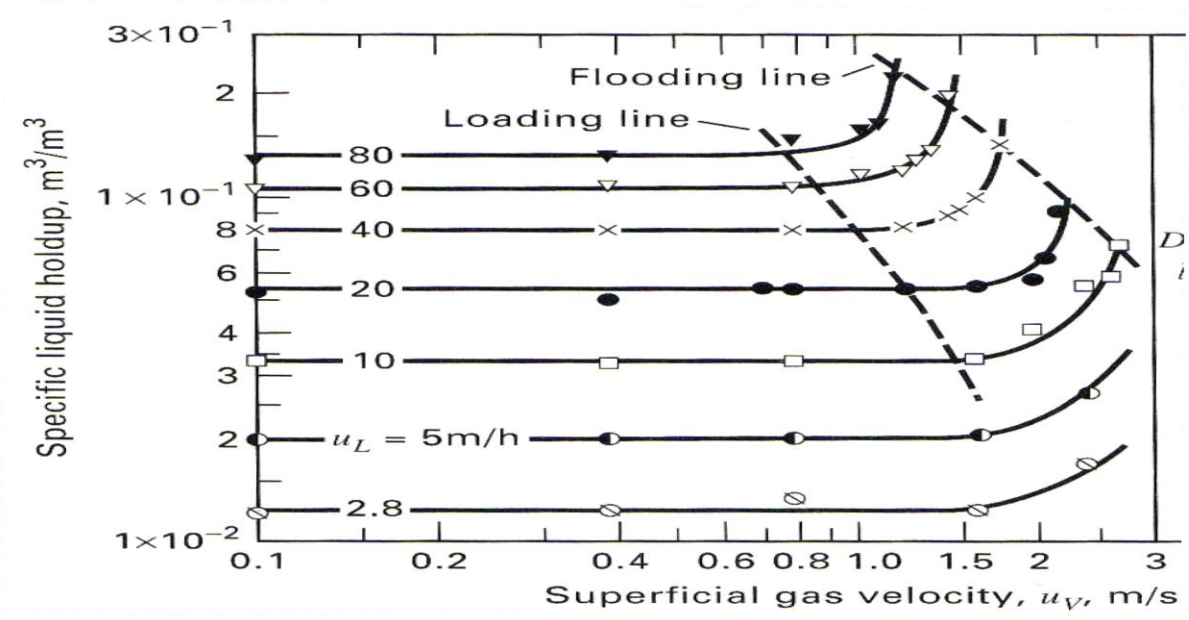
A number of parameters have to be fixed before the diameter of the column can be calculated. These include fluid molar flows and physical properties as well as the packing factor of packing in the tower. Nevertheless, it is important to discuss the pressure drop in the packed tower as the diameter calculation is intertwined with it. (Green & Perry 2008, 14-9; McCabe et. al. 2005, 576.)

Experiments conducted by Billet, and also Stichlmair, Bravo and Fair to investigate the variation of pressure drop and liquid holdup in the column at different super-

ficial gas velocity, the velocity of gas phase in an empty column, yield the useful graphs presented. (Seader & Henley 2006, 228.)



GRAPH 6. Pressure drop characteristics of packed columns (McCabe et. al. 2005, 570)



GRAPH 7. Specific liquid holdup chart of packed columns (Seader & Henley 2006, 228)

From Graph 6 it can be observed that a linear relationship exists between the pressure drop and superficial gas velocity. More accurately, the pressure drop is proportional to the velocity to the power of 1.86 when liquid flow is not present. On the introduction of the liquid flow, the pressure drop increase is higher compared to the dry case. This increase is attributed to liquid holdup in the column which reduces the void space available for the gas throughput by its magnitude. The liquid holdup as seen from Graph 7 is constant over a certain range. The pressure drop in this range maintains the linearity proposed earlier. This range of constant liquid holdup in the column is termed the preloading region, and here, the gas phase is the continuous phase. A set of equations presented by Billet and Schultes involving two dimensionless parameters, Reynolds and Froude number, can be used to estimate the liquid holdup in the preloading region. (Green & Perry 2008, 14-56—14-57; McCabe et. al. 2005, 569-571; Seader & Henley 2006, 228-229; Zenz 1980, 289-307.)

Above the preloading region, the pressure drop and liquid holdup cease to be constant because of liquid accumulation in the column. This continues until the liquid phase becomes the continuous phase, usually at about 2.0 in. water per ft. packing in most packing. The incipient of liquid accumulation in the unit is termed the loading point, whereas when the liquid phase just dominates is termed the flooding point. Flooding in the column is recognized by a sharp increase in pressure drop, liquid holdup, appearance of liquid on top of the bed, and a drop in mass transfer efficiency. At this very stage and above, the liquid phase domination hinders the movement of the gas phase up the tower. The region between the loading point and when the column starts to flood is referred to as the loading region. The most efficient condition is reached in between the loading region; thus, design calculations are made to fall within it. Similar graphical representations as

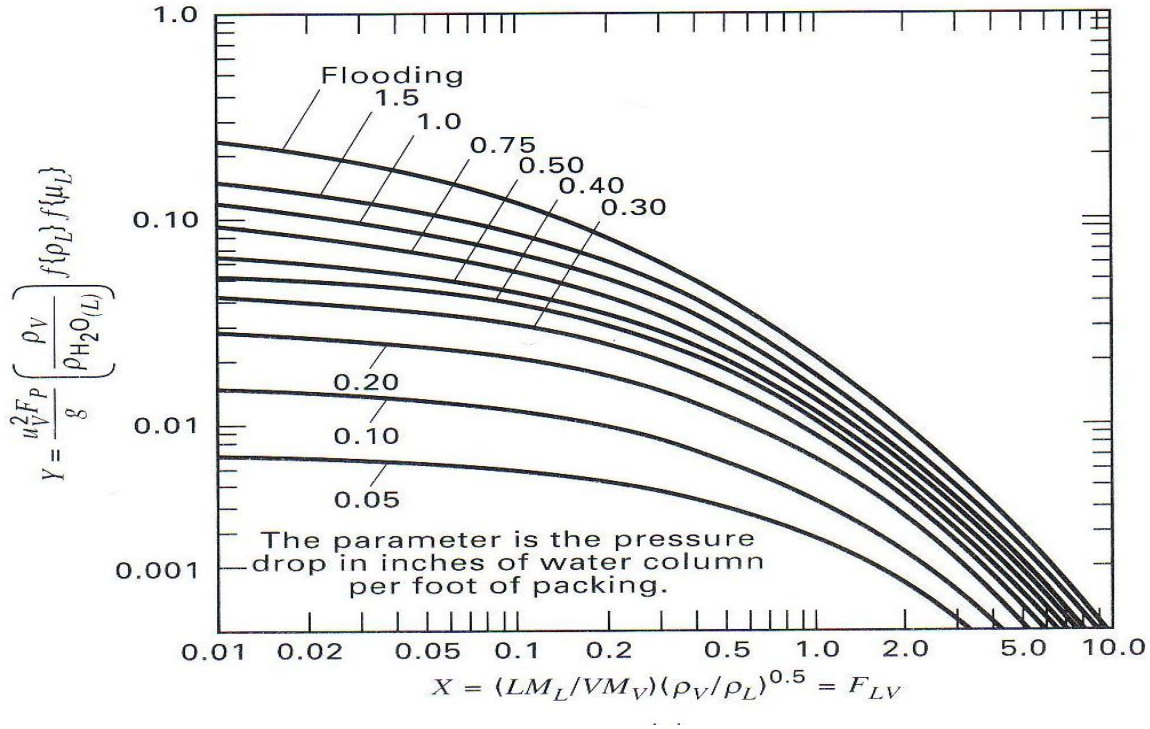
in Graphs 6 and 7 can be arrived at with different packings and fluids. (Green & Perry 2008, 14-56—14-57; McCabe et. al. 2005, 569-571; Seader & Henley 2006, 228-229; Zenz 1980, 289-307.)

The perceived dependency of flooding conditions on the fluids being handled and the packings characteristics prompted an attempt to develop a general correlation graph for the flooding data; particularly superficial gas velocity and pressure drop. Sherwood, Shipley and Holloway who are the pioneers in this attempt came up with the first generalized flooding-data correlation in 1938 (Seader & Henley 2006, 233-236; Zenz 1980, 289-307). Later, Leva improved this chart and added some constant pressure drop lines which are the origin of the name generalized pressure-drop correlation (GPDC) (McCabe et. al. 2005, 569-575; Seader & Henley 2006, 233-236). In Eckert's version, a flooding line located above 1.5in. water per feet packing was removed due to its inconsistency his experimental studies; still further adjustment was made by Stringle to both axes to make better the correlation. Even with the tenths of improvements, Kister and Gill noted that structured packings yield steeper curves, so they developed a special one for them (Green & Perry 2008, 14-55—14-62; McCabe et. al. 2005, 569-575). The accuracy of the predicted pressure drop with the GPDCs is still questionable especially above 50% of flooding velocity. Thus, Kister & Gill and Stigle suggest the use of the equation below in recognition of the influence of packing factor on the pressure drop at flooding point

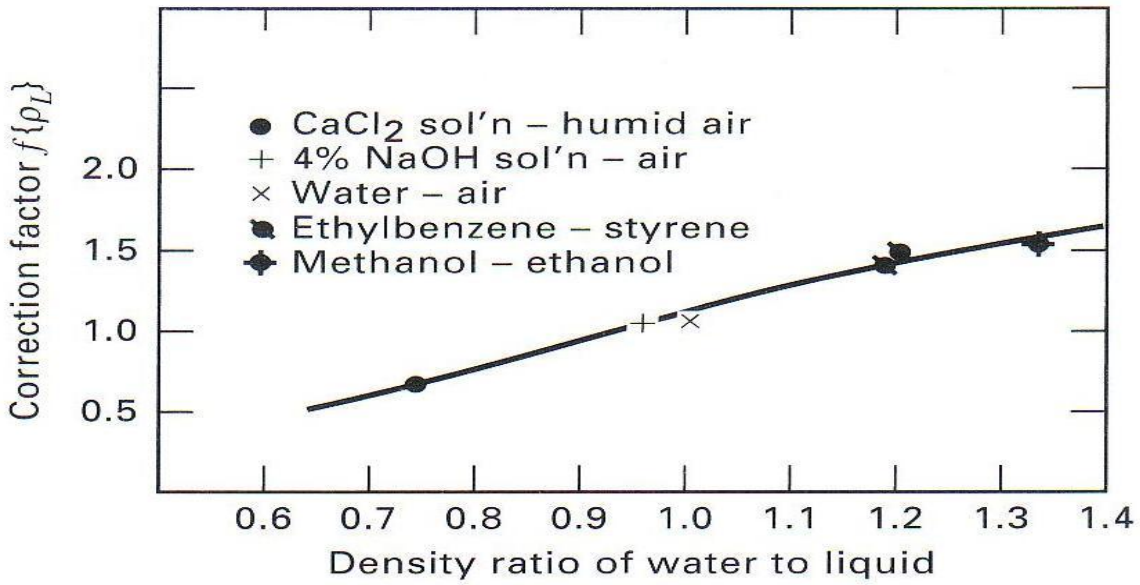
$$\Delta P_{flood} = 0.115 F_p^{0.7}$$

where ΔP_{flood} is the pressure drop at flooding in inches of water per feet of packing, and F_P is a dimensionless parameter called the packing factor. A number of other researchers have constantly tried to improve the correlation data. Notable versions of GPDC and/or empirical equations exist that are accredited to the following researchers: Robbins; Mackowiak; Eiden & Bechtel and Lockett & Billing-

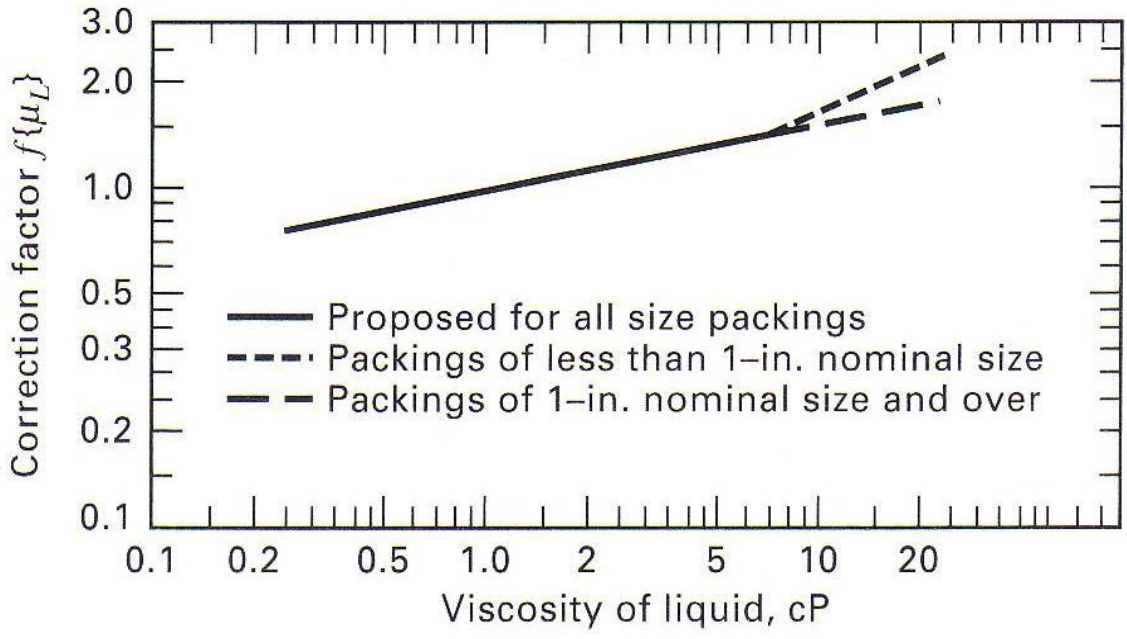
ham. Furthermore, in 1992 Leva published a modern version of his GPDC chart with a new y-axis that includes a correlation factor for the viscosity and density of the liquid in use. Graphs 8, 9 and 10 represent Leva's latest GPDC chart alongside the viscosity and density correlation graphs. (Green & Perry 2008, 14-55—14-62; McCabe et. al. 2005, 569-575; Seader & Henley 2006, 233-236; Zenz 1980, 289-307.)



GRAPH 8. Leva's generalized pressure-drop correlation (Seader & Henley 2006, 233)



GRAPH 9. Liquid density correction chart (Seader & Henley 2006, 233)



GRAPH 10. Liquid viscosity correction chart (Seader & Henley 2006, 233)

With the necessary fluids parameters and packing factor available, the ordinate parameter of the GPDC chart can be calculated. Using the calculated value and the flooding curve, the corresponding abscissa magnitude can be detected. Utilizing

this value hand in hand with Graphs 9 and 10, the superficial gas velocity at flooding can be determined and then the column diameter at the required percent of flooding can be calculated with the equation below

$$D_T = \left(\frac{4VM_V}{fu_{V,f}\pi\rho_V}\right)^{0.5}$$

Theoretical approaches to packed column analysis by Stichlmair, Bravo & Fair (particle model); and Rocha, Bravo & Fair; Mackowiak; and Billet & Schultes (channel model) all came up with well acceptable equations (Green & Perry 2008, 14-55—14-62; Seader & Henley 2006, 228-237). In 1999 Billet & Schultes presented another model, semi theoretical this time but probably more accurate, with which superficial velocity, pressure drop, and column diameter can be calculated both at the loading and flooding point. With the following set of equations, the aforementioned parameters at flooding can be determined (Seader & Henley 2006, 228-237)

$$\frac{\Delta P_o}{l_T} = \psi_o \frac{a}{\epsilon^3}\frac{u_V^2\rho_V}{2}\frac{1}{K_W}$$

where ΔP_o is the pressure drop at zero liquid flow, l_T is the height of packing, K_W is the wall factor, ψ_o is an empirical constant, u_V denotes the superficial gas velocity, whereas ρ_V means the gas density, a, and ϵ represents the interfacial area and the porosity of the packings respectively

$$\frac{1}{K_W} = 1 + \frac{2}{3}\left(\frac{1}{1-\epsilon}\right)\frac{D_P}{D_T}$$

where D_P and D_T are the effective packing diameter and the tower's diameter sequentially

$$D_P = 6\left(\frac{1-\epsilon}{a}\right)$$

The empirical constant and the Reynolds number, N_{Re_V}, can be evaluated with the equations below

$$\psi_o = C_P\left(\frac{64}{N_{Re_V}} + \frac{1.8}{N_{Re_V}^{0.08}}\right) and\ N_{Re_V} = \frac{u_V D_P \rho_V}{(1-\epsilon)\mu_V}K_W$$

Lastly, the pressure drop at flooding, ΔP, is given by this

$$\frac{\Delta P}{\Delta P_o} = \left(\frac{\epsilon}{\epsilon - h_L}\right)^{3/2} exp\left[\frac{13300}{a^{3/2}}\left(N_{Fr_L}\right)^{1/2}\right]$$

The liquid holdup and the Froude number can be calculated with the mathematical relations below:

$$h_L = \left(12\frac{N_{Fr_L}}{N_{Re_L}}\right)^{1/3}\left(\frac{a_h}{a}\right)^{\frac{2}{3}}$$

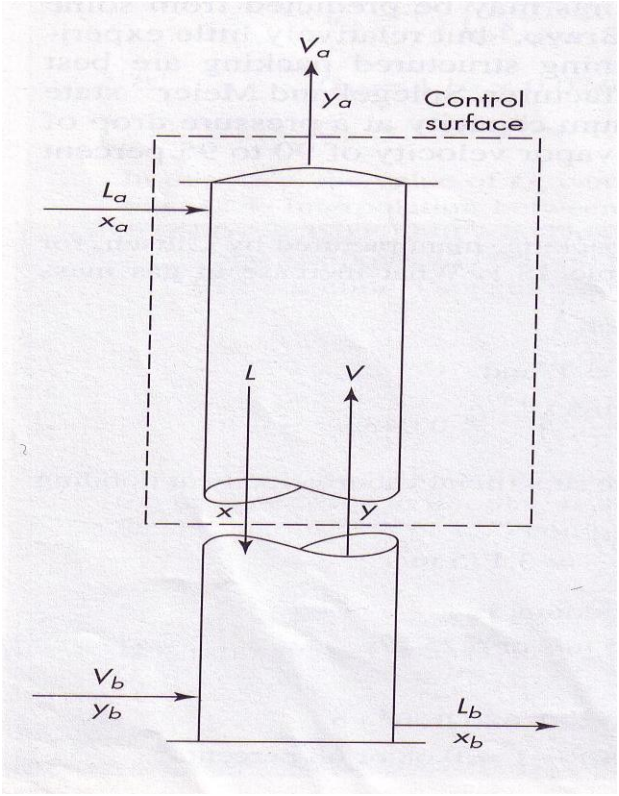
$$N_{Fr_L} = \frac{u_L^2 a}{g} \;\; and \; N_{Re_L} = \frac{u_L}{a v_L}$$

$$a_h/a = C_h N_{Re_L}^{0.15} N_{Fr_L}^{0.1} \;\; when \; N_{Re_L} < 5$$

$a_h/a = 0.85 C_h N_{Re_L}^{0.25} N_{Fr_L}^{0.1} \;\; when \; N_{Re_L} \geq 5$. (Green & Perry 2007, 14-55—14-62; Seader & Henley 2006, 228-237.)

The height of packings is probably the last task to be resolved in the design of a packed column. The amount of the target solvent absorbed, the mass transfer efficiency of the equipment, and the prevailing equilibrium condition all work together to determine the craved height. (Green & Perry 2008, 14-9; McCabe et. al. 2005, 576; Seader & Henley 2006, 223-227; Zenz 1980, 289-307.)

In view of the continuous nature of the packings arrangement in the column, the use of differential equation is inevitable in its analysis. In Graph 11, the material balance would be considered both around the dashed section and an infinitesimal area of the column with height dZ to develop the equation for the operating line and the height of the column respectively.



GRAPH 11. Sketched packed column showing the streams (adapted from McCabe et. al. 2005, 576)

The component material balance for the absorbed gas around the area marked with dash lines yields the following

$$L_a x_a + Vy = Lx + V_a y_a$$

Reshuffling this to express gas composition, y, in terms of the liquid phase composition, x, the general equation describing the operating line is arrived at as shown below:

$$y = \frac{L}{V}x + \frac{V_a y_a - L_a x_a}{V}$$

In these equations, L and V denotes the liquid and vapour molar flow, while x and y stands for the mole compositions of the solute in the liquid and gas phase respectively. Likewise, the material balance through the infinitesimal area assuming a dilute solution (less than 10% mole composition as rule of thumb) leads to this

$$-Vdy = K_y a(y - y^*)SdZ$$

where K_y is the overall gas-phase mass transfer coefficient, y^* represents the gas mole composition that will be in equilibrium with x; S is the cross sectional area of the column and dZ the height of the infinitesimal portion.

Rearranging and solving this mathematical relation for the height Z gives

$$Z = \frac{V/S}{K_y a}\int_a^b \frac{dy}{y - y^*}$$

This equation contains two distinct parts; $\frac{V/S}{K_y a}$ and $\int_a^b \frac{dy}{y-y^*}$. The former is termed the overall height of a transfer unit, HTU, and the latter is the number of transfer unit, NTU, based on the gas phase. The HTU is a measure of the mass transfer effectiveness of the equipment, the smaller the value the better the equipment. It can be seen from the expressions above that HTU varies directly with the gas molar flow and inversely with $K_y a$. HTU's variation is less pronounced with changing vapour flow as compared to $K_y a$. In order to calculate HTU from the expression above, $K_y a$ has to be estimated. The estimation of $K_y a$ theoretically is a broad topic whose discussion is out of the scope of this thesis. Also, it should be noted that the values gotten theoretically usually show significant deviation from the ones gotten from experiment. An accurate determination of HTU relies on the experimental data from the packing's manufacturer. This can be a graphical plot of HTU against a flow parameter whose equation would be provided. The magnitude of HTU and $K_y a$ are not a constant. They change with different fluids throughput. (Green & Perry 2008, 14-11—14-13; McCabe et. al. 2005, 580-584; Seader & Henley 2006, 223-227.)

The NTU is a dimensionless number that gives an insight into the difficulty encountered in effecting the required separation. It is regarded as the difference in the solute mole composition per average driving force in mole fraction. High NTU values indicate purer outlet gas. The definite integral above can be solved to get

the NTU value or better still the use of the logarithmic mean average so as to do without integrating; the equations needed in both cases are presented below.

1. Logarithmic mean:

$$NTU_{OG} = {y_b - y_a}/{(y - y^*)_{LM}}$$

$$(y - y^*)_{LM} = \frac{(y_b - y_b^*) - (y_a - y_a^*)}{\ln\left[\frac{(y_b - y_b^*)}{(y_a - y_a^*)}\right]}$$

2. Integral method: The first solution to the definite integral was provided by Colburn in 1939. He assumed a linear equilibrium and operation lines in order to simplify the equation as thus:

   Equilibrium curve equation: $y^* = Kx$

   Operating line equation: $y = \frac{L}{V}x + \frac{V_a y_a - L_a x_a}{V}$, note Va ≅ V and La ≅ L because of the dilute assumption.

   With these equations, the integral can be resolve as thus;

$$\int_a^b \frac{dy}{y - y^*} = \int_a^b \frac{dy}{(1 - KV/L)y + y_a(KV/L) - Kx_a}$$

   When A = L/(KV), the following expression was arrived at

$$NTU_{OG} = \ln\left\{\left[{(A-1)}/{A}\right]\left[{(y_b - Kx_a)}/{(y_a - Kx_a)}\right] + {1}/{A}\right\} \Big/ {(A-1)}/{A}$$

When dealing with concentrated solution, probably with solutes greater than 10% as a rule of thumb, also when using a chemically absorbing solvent, the assump-

tion which simplifies the integration process does not hold anymore because both the equilibrium and or the operating line cease to be linear. In such a situation, numerical integration has to be performed on the unsplitted definite integral derived for the calculation of the required height of packings with all the parameters, except the cross sectional area, included in the integral. (Green & Perry 2008, 14-11—14-13; McCabe et. al. 2005, 576-600; Seader & Henley 2006, 223-244.)

3 DESIGN OF EXPERIMENT

An experiment refers to a vital investigative tool utilized in the sciences to either validate or disapprove a proposed hypothesis. Put in another way, experiments are used to investigate the influence of changes in independent variables on the dependent one(s) under inspection. Experimentation has proven to be a very useful tool to the academic world, especially in sciences and engineering where a large number of empirical equations exist even when little or nothing is known about the theory behind them. The industry is not left behind as regards the benefits of experimentation. Feats such as product optimization, and cost minimization cannot be adequately done without proper understanding of the variables affecting the productivity in question. (Experiment 2010; Statsoft 2010.)

The design of experiment is a branch of statistics that deals with planning and carrying out of experiments, and interpretation of the results. The subject matter of design of experiment is twofold: one is how many runs will be made, and the other is in what condition will the runnings be made, that is, the ways the factors are put together to extract unbiased information (Roy 1990, 44). Following this, is the mathematical analysis of the results obtained from the tests conducted. As regards the experimental plan, a number of established approaches are available for the experimenter to choose from depending on his aim and size of variables. In academic research, where less parameter are studied and high accuracy of the result matters, the traditional factorial design which tests all the possible combinations is suitable. In the industry, however, a large number of variables will commonly affect productivity plus the fact that resources and cost minimization are usually on

top of the scale of preference. Thus, industrialists instead sought techniques to better cut cost and resources, besides the factorial approach is impractical when large number of parameters affect production as is always the case in a typical industrial setting. (Statsoft 2010; Roy 1990, 1-5.)

Following the proposal of the factorial design approach to experiment design in the 1920s by Fischer, a handful of other methods have been developed by other statisticians to suit the industrial need (Roy 1990, 40-41). However these other proposals were in ink, their applications are still seldom due to ambiguity in quality definition. In the process of reducing costs on experimentation while resolving the Japanese telecommunication problem, Dr. Genichi Taguchi, came up with a new philosophy coupled with his redefinition and treatment of quality in a way appealing to the industry (Roy 1990, 7). The totality of his approach is referred to as the Taguchi method. Adherence to this approach has helped the Japanese manufacturing industry attain the quality which they are known for today. However, it is not until in the 1980s that the western companies begin to see the beauty of this novel system. AT&T and Ford Motor Company are amongst the first American companies to testify to the quality improvement attained with the use of Taguchi's method (Statsoft 2010).

3.1 The Taguchi approach to quality improvement

Taguchi's philosophy towards achieving quality improvement can be put down in three simple statements: Firstly, provision for quality at the primary level, that is, quality should be catered for at the planning stage. It is better to tackle the quality

problem at the start rather than checking products at random when the fault is already made. Secondly, quality should be quantified as nearness to the target. In other words, the aim should always be the target and not a specific range around the target as it is in quality checking method. Thirdly, expenses incurred in attaining quality should be expressed in terms deviation from target. This cost estimating function is commonly known as the lost function. (Roy 1990, 8-10.)

The totality of the Taguchi approach to quality engineering was born out of the three simple concepts discussed above. The central message of Taguchi's philosophy is that quality is a measure of proximity to the set standard, and consistency of individual trials. A three-level design is recommended for good quality; systems design, parameter design and tolerance design. The strategies toward achieving this quality are as follows: Firstly, optimization of the whole process; secondly, making the system robust, less responsive to noise—uncontrollable factors—variables, without eliminating the noise source. The two strategies are made possible through the use of experimental procedures as proposed by the Taguchi's method. (Roy 1990, 10-28.)

The quality characteristics are the property of a product through which the level of attainment of the desired quality can be identified. The quality characteristics of most process or product can be expressed numerically while others are not. Also, more than one characteristic can determine the quality of a product; say a casted metal where the lustrousness and malleability are the quality determinant. In the case of multiple quality characteristics, a single quality property called overall evaluation criteria (OEC) must be deduced with the mathematical relation below.

$$OEC = \left({}^{y_1}/_{y_{1max}}\right) \times w_1 + \left({}^{y_2}/_{y_{2max}}\right) \times w_2 + \cdots$$

Here, w_i = weight of the ith component, y_i = measurement of the ith criterion; and y_{imax} = maximum value of ith criterion. The weights of the components would be decided in the brainstorming session which would be discussed later on. The y values however would be gotten from the experimental data.(Roy 1990, 178-179.)

Whatever kind of quality characteristics a product possess, the Taguchi method has three categories, listed below, in which the product's quality criteria must fall. (Roy 1990, 19-20.)

1. The bigger the better: This is applicable to good characteristic which is desired to be maximized
2. The smaller the better: Applicable to undesirable characteristic which is to be minimized
3. The nominal is best: This is the case when a known value is the focus.

3.2 Designing experiment the Taguchi way

It has been emphasized earlier that quality is equivalent to proximity to target, according to the Taguchi method, and also that the strategy towards achieving this is to optimize the process, or perhaps tune the parameters in such a way that the noise parameters are less dominant on the product. Of the three levels of design to be followed, parameter design is relevant to determining the optimum variables combination that gives the desired goal. Discussing the application of process and tolerance design is out of the scope of the design of experiment.

Based on the Taguchi approach to experiment design, the search for the right combination of parameters that will yield the optimum result is a four-step process, each of which will be discussed in the subsequent headings.(Roy 1990, 29.)

3.2.1 Brainstorming

Brainstorming is the starting point and the most crucial step in the design of experiment. It is a meeting session that involves the participation of staffs from various departments with the aim of deciding the conditions of the experiments to be ran. It is encouraged to involve as much departments there are that influence the quality of the product. Thus the design would be made better as two heads are better than one. Common issues talked about in a brainstorming session are as follows: What factors are affecting the products and what are their levels? Which factors and levels should be involved in the study? What are the noise factors available? Are there interactions, if so should they be studied? What is/are the quality characteristic(s)? In the case of multiple quality parameters, how are they to be combined/ what is each component's weight? In what category does the quality characteristics belong? (Roy 1990, 29 & 173-179.)

3.2.2 Designing and executing

Having held the brainstorming session and after reaching consensus on all the important questions raised during the meeting, the next thing is to design and execute the experiments. Designing an experiment is synonymous to determining the number of runs and the condition in which the runnings will be made. Taguchi proposed the use of standard orthogonal arrays for designing experiments. The standard orthogonal array is a tabulated set of experimental conditions extracted from a whole list of the conditions offered by the traditional factorial approach to experiment design. The selected set of conditions presented in the orthogonal array is the minimum through which the same inference drawn in the factorial's list can be made. Orthogonal arrays are carefully constructed to yield unique design to the same situation by different experimenter; also, the results obtained are susceptible to statistical analysis. Tabular representation, such as Graph 12 below, from which the needed standard orthogonal array can be looked up, abounds in the literature. (Roy 1990, 29-30.)

		Number of Parameters (P)												
		2	3	4	5	6	7	8	9	10	11	12	13	14
Number of Levels	2	L4	L4	L8	L8	L8	L12	L12	L12	L12	L16	L16	L16	L16
	3	L9	L9	L9	L18	L18	L18	L18	L27	L27	L27	L27	L27	L36
	4	L16	L16	L16	L16	L32	L32	L32	L32	L32				
	5	L25	L25	L25	L25	L25	L50	L50	L50	L50	L50	L50		

GRAPH 12. Orthogonal array selector (adapted from DOE 2007)

The number in the names given to the standard orthogonal arrays denotes the number of test runs required for the specific situation. As an illustration, an experimenter with four parameters with three levels each at hand will make use of array L9 according to Graph 12 above. Presented below is the layout of the conditions embodied by the L9 array.

Experiment	P1	P2	P3	P4
1	1	1	1	1
2	1	2	2	2
3	1	3	3	3
4	2	1	2	3
5	2	2	3	1
6	2	3	1	2
7	3	1	3	2
8	3	2	1	3
9	3	3	2	1

GRAPH 13. L9 orthogonal array's layout (adapted from DOE 2007)

With the right array at hand, experiments are designed by mimicking the array. Once the design is completed, the experiments are run accordingly. Issues regarding execution of the experiments are usually process dependent, for example, the order in which the runs are made relies on the ease of tuning the parameters' levels. (Roy 1990, 40-47.)

3.2.3 Analysis of results

The process of analyzing the result from an experiment follows a definite pattern towards achieving its aims in Taguchi's philosophy. The aims of the analysis are 1. to determine the optimum condition, that is, to recognize the set of variable that yields the result nearest to the target; 2. to estimate the expected performance at the optimum condition; and 3. to determine the influence of each of the factors on the results. (Roy 1990, 47-48.)

The analysis process can either make use of the average or signal to noise ratio values. Apart from the calculation of the averages or the signal to noise ratios, the analysis procedure is basically the same irrespective of the quantity adopted. When repetitions of runs are carried out, Taguchi recommends the use of his signal to noise ratio formulae for the analysis of the results. Otherwise, the conventional averages are made use of. The transformation of the results to signal to noise(S/N) ratio is made possible with the formula stated below

$$S/N = -10 \times \log_{10}(MSD)$$

Where MSD is the mean squared deviation from the target. Mathematical expressions for the computation of the MSD for the quality characteristics discussed in earlier heading are as follows:

Smaller the better: $MSD = {(y_1^2 + y_2^2 + y_3^2 + \cdots)}/{n}$

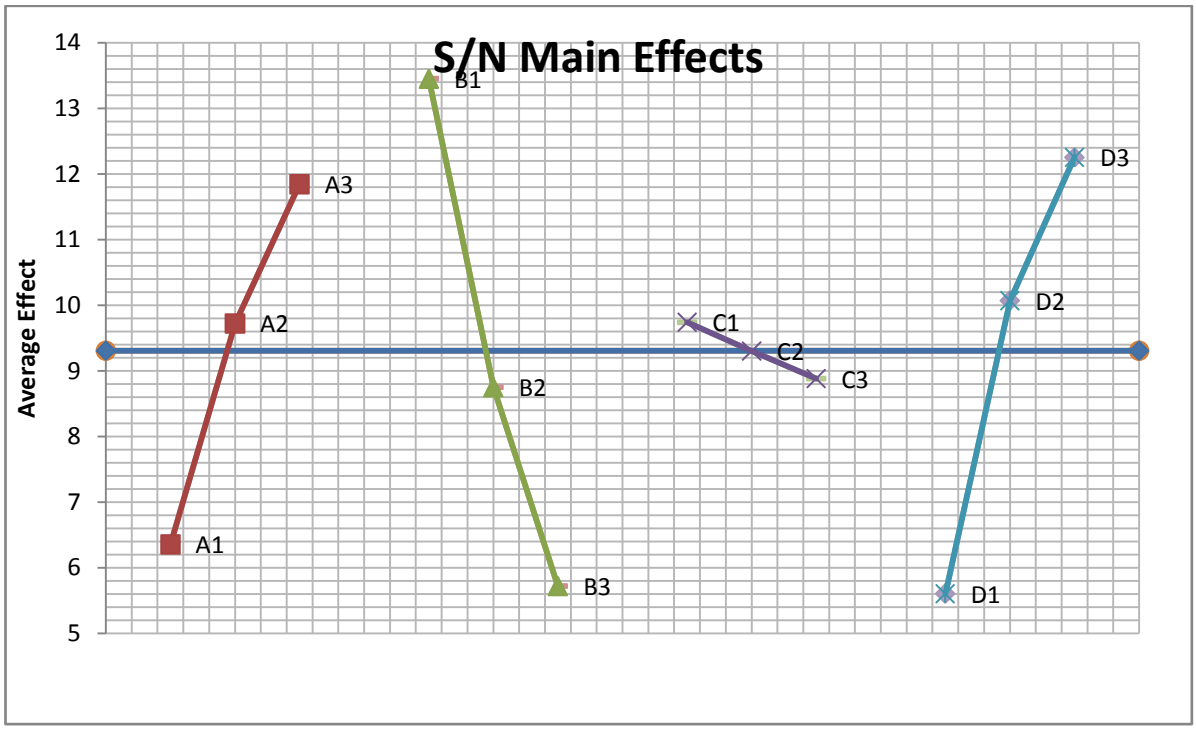
Nominal is best: $MSD = {((y_1 - m)^2 + (y_2 - m)^2 + \cdots)}/{n}$

Bigger the better: $MSD = \left({}^{1}/_{y_1^2} + {}^{1}/_{y_2^2} + {}^{1}/_{y_3^2} + \cdots\right)\Big/_{n}$

In these equations, y1, y2, y3 are the results for a particular run at the first, second and third repetitions respectively; m connotes the target value and n is the number of repetitions. With the aid of the appropriate formula, the signal to noise ratios for all the trial conditions in an experiment can be calculated. If the averages were employed, however, the average value of the n repetitions for every trial condition would be computed instead. Whichever is the case, either with the signal to noise ratios or the average values, the average effect of all the studied parameters are calculated at their respective levels. The average effect of a parameter at a specified level is accounted for by averaging the results of all the tests containing the parameter at the level in question. (Roy 1990, 47-98.)

Graphical depiction of average effects of parameters against their respective levels, for example, as shown in Graph 14 below, is a necessity for the determination of the optimum state. This graphical illustration is commonly referred to as the main effects plot. The horizontal line running through the plots is the grand average of the whole results. This graph makes visible the main effects, the difference between the average influences of each variable. Thus, preliminary guesses can be made about the factors influence on the product. The same scale is applied to all parameter plotting so as to facilitate comparation. The optimum combination is easily recognized from the graphical illustration. The desired variable combination depending on whether the quality characteristics are bigger the better, smaller the better or nominal the best is the factors' level-set at the top, bottom or nearest to the target respectively. The optimum conditions in Graph 14 below are A1B3C3D1

and A3B1C1D3 for smaller the better and bigger the better sequentially. (Roy 1990, 47-98.)



GRAPH 14. A typical main effects plot.

Having achieved the optimum set of variables, the expected performance can be estimated with these simple expressions assuming the cases, smaller the better and bigger the better in Graph 14 above:

$Y_{opt} = GA + (A1 - GA) + (B3 - GA) + (C3 - GA) + (D1 - GA)$ and

$Y_{opt} = GA + (A3 - GA) + (B1 - GA) + (C1 - GA) + (D3 - GA)$ where GA is the grand avergae of all the processed results from the experiment. Further inquiry into the factors' influence on the products can be made with the famous analysis of variance (ANOVA). It should be noted that factors contributions calculated with the ANOVA technique are relative to one another. The result of the ANOVA

calculation is usually presented in a table showing a number of calculated quantities and the relative contribution of each factor in percent. Table 1 below shows an ANOVA table with all its presented quantities usually calculated. (Roy 1990, 47-98.)

TABLE 1. An ANOVA table

COLUMN	FACTORS	DEGREE OF FREEDOM	SUM OF SQUARES	VARIANCE	VARIANCE RATIO	PERCENT CONTRIBUTION
1	Factor1	--	--	--	--	--
2	Factor2	--	--	--	--	--
3	Factor3	--	--	--	--	--
Others/Error		--	--	--	--	--
Total		--	--			100

The quantities shown in Table 1 above are briefly explained below with the appropriate formula for their evaluation when necessary:

- Degree of freedom: The degree of freedom, DOF, is one less than the number of levels of a variable. Similarly for an array, the DOF is one less than its number of levels or rows it contains. Thus a parameter with three different levels has two as its DOF, whereas an L9 array displayed in Graph 2 has eight as its DOF. However, the DOF for a set of experimental result is calculated as follows:

  DOF = number of results - 1;

and DOF = number of results *number of repetition – 1, when there is repetition. The first expression is always the case when S/N is used in the analysis process. Lastly, the DOF for error variance is that of the total experiment set minus that of each of the variables in the experiment.

- Sum of squares: The total sum of squares, S_T, is calculated by summing the squares of the results of the experiment and subtracting the correction factor, CF, from it. The equations are presented below

$$S_T = \sum y^2 - CF \;\; where\; CF = {(\sum y)^2}/{n}$$

The factors' sum of squares can be calculated as illustrated below using a factor A with two levels as an example

$S_A = {A_1^2}/{N_{A1}} + {A_2^2}/{N_{A2}} - CF$ where A_1 and A_2 are the sum of the results in which level1 and level2 values of the factor A are present; and N_{A1} and N_{A2} are their respective number of experiments in which factor A's level 1 and 2 participated or the number of results summed up to get A_1 and A_2. Just like with the DOF, sum of squares for error is the total sum of squares minus those of the factors. The estimation of the other error parameters takes the same approach.

- Variance: The term variance is defined as the ratio of the sum of squares to the DOF. For example, factor A's variance can be calculated as $V_A = {S_A}/{DOF_A}$.

- Variance ratio: This is the ratio of the variance of any factor to that of the error. When the error's variance is zero, the variance ratios are all rendered indeterminate. In such a case, the error's variance can be combined with

that of any factor whose value is relatively insignificant. Say a variable B possesses small variance, then that of the error can be reestimated as thus; $V_{error} = {}^{S_B + S_{error}}/_{DOF_B + DOF_{error}}$. The merging of any factor's variance with that of the error is termed pooling. Afterwards, the variance ratios can be calculated and the sum of squares are also adjusted as follows; $S_A^{'} = S_A - (V_{error} \times DOF_A)$. The newly calculated sums of squares- pure sum of squares- are used for the contribution calculation.

- Percentage contribution: The ratio of the sum of squares of a factor to that of the total result multiplied by hundred is called the factor's contribution in percent. The percentage contribution or influence of the factors on the result is that goal of the ANOVA calculation

$$P_A = {}^{S_A}/_{S_T} \times 100$$

(Roy 1990, 47-98 & 101-155.)

3.2.4 Confirmatory test

Running a confirmatory test is usually the last stage in the process of experimenting following the Taguchi's approach. As a continuation from the results analysis, the optimum result, Y_{opt}, predicted at the determined optimum condition must be verified. This is actually one of the beauties of the Taguchi's method. In order to validate the prediction, new experiment trials, whose results will be juxtaposed with the one arrived at by equation, have to be made. (Roy 1990, 31; Statsoft 2010.)

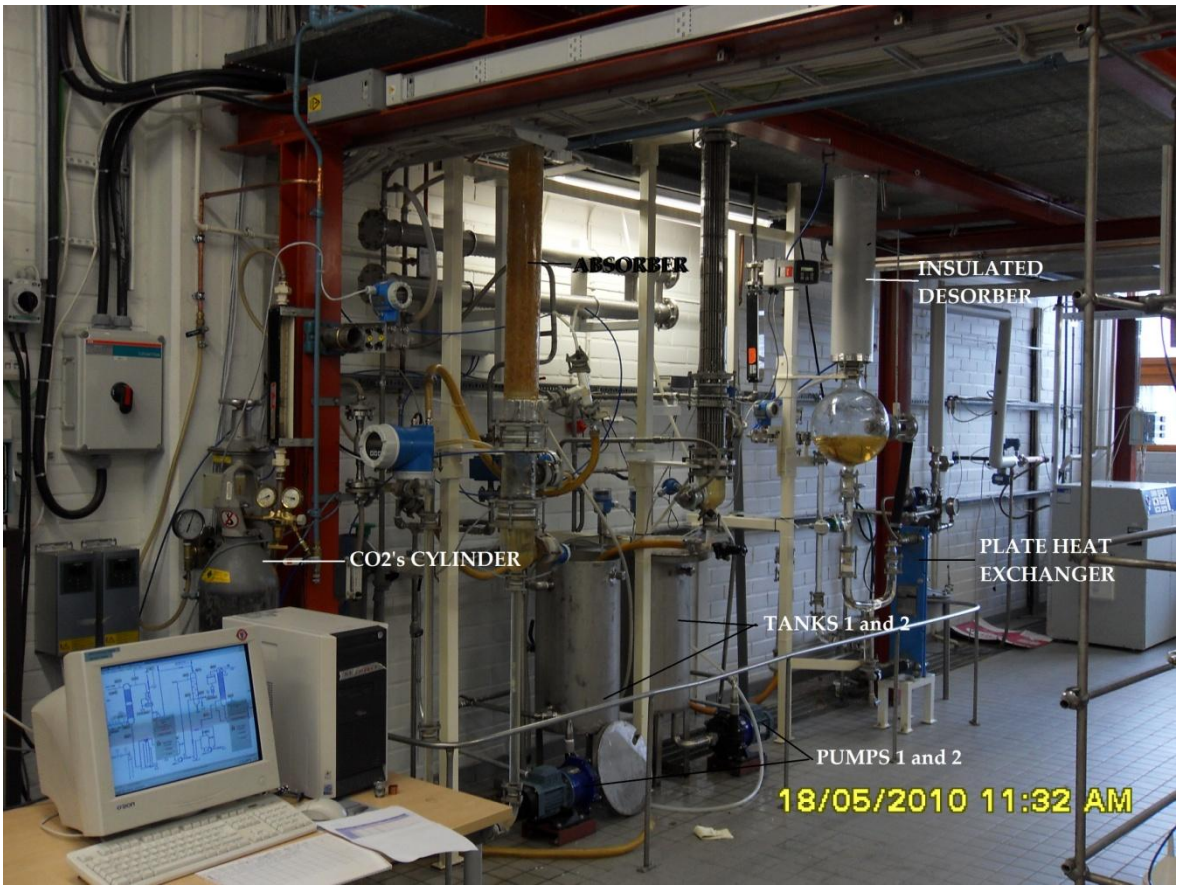
The comparation of the two results should show a reasonable agreement; otherwise possible interaction between variables might have prompted a disagreement. The decision of whether or not to study interactions is made in the brainstorming stage. If an interaction is left out when it is significant, it will tell on the final results; this further emphasizes the need for an elaborate brainstorming session.

4 EXPERIMENTATION

The pilot scale CO_2-MEA absorption-desorption unit installed in the laboratory of Central Ostrobothnia University of Applied Sciences was observed to accumulate CO_2 as it was being run. It was also observed that this was as a result of the inefficiency of the desorber which constantly floods. Thus, this experiment is aimed at optimizing the CO_2 removal or regeneration of the washing liquid in the stripper. This experimentation will follow closely the Taguchi approach, and thus, the proposed four-stage process for the determination of the optimum condition would be strictly adhered to. A detailed description of the experiment equipment and procedure will be offered in the subsequent subheadings.

4.1 Equipment description

The CO_2-MEA absorption-desorption unit utilized in this experiment, depicted in Graph 15, has the following features: an absorber of diameter 100mm and a desorber of diameter 80mm both made of transparent glass and packed with 10mm Raschig ring; a shell and tube heat exchanger positioned in between the packed columns for allowing heat exchange between the two columns feed; two storage tanks with a centrifugal pump each for the circulation of the washing liquid; a plate heat exchanger through which hot steam's flow energizes the stripper's content to release CO_2 and steam; a heating coil for preheating the stripper's feed, and a metallic cylinder which contains CO_2 gas.



GRAPH 15. The CO_2-MEA absorption-desorption unit

The control and measurement facilities of the parts of the unit mentioned are pinpointed below:

- The pumps are controlled from the user interface on a desktop computer. The pumps run based on the set flow rate and they also work in such a way that the liquid levels in the tanks are always leveled. The tank level can be seen from the user interface.
- The carbon dioxide flow can be adjusted with the knob on the cylinder while the airflow is controlled with a valve linked with the pipeline supplying air to the laboratory. The two streams are mixed on their way to the absorber where an online sensor has been installed to detect the CO_2 content.

- The preheater is the easiest to control. It has three power levels which can be chosen with a switch.
- The steam flow can be controlled by turning a restrictor either clockwise or anticlockwise. As the restrictor is being turned, the corresponding pressure can be checked on a pressure meter installed along the steam's passage.
- Lastly, several online sensors were installed at different points within the columns to measure pressure drops and temperatures.

4.2 Experimental procedure

In line with the principles of parameter design, the experiment started with the brainstorming session which had five participants: the project supervisor, laboratory assistance, and three students whose theses are related to the subject matter. The aim of the meeting, which was to optimize the CO_2 removal in the desorber, was stated and then exchange of ideas began on how to achieve the goal. In about an hour of discussion, concessions were reached as regards the factors affecting the functionality of the stripper column to be considered in the experiment, the levels of variables to be tested; the performance parameter, and the quality characteristics. The agreement reached is summarize in Table 2 below.

TABLE 2. Agreed condition for the MEA regenerating process optimization

Variables	Level 1	Level 2	Level 3
CO_2 load (%)	10	15	20
Liquid flow (l/min)	2.5	3.0	3.5

Steam's pressure (bar)	0.20	0.25	0.30
Preheater's power (kW)	2	4	6
Performance parameter : CO_2 removal in the stripper			
Quality characteristics : Bigger the better			

Following the accord on the variables and levels to be studied, a L9 array was used to design the experiment. Three repetitions were made each so as to use the signal to noise ratio in the analysis. Table 6 below shows the experiment conditions as deduced with the L9 array.

TABLE 3. Experimental condition layout for MEA regeneration optimization

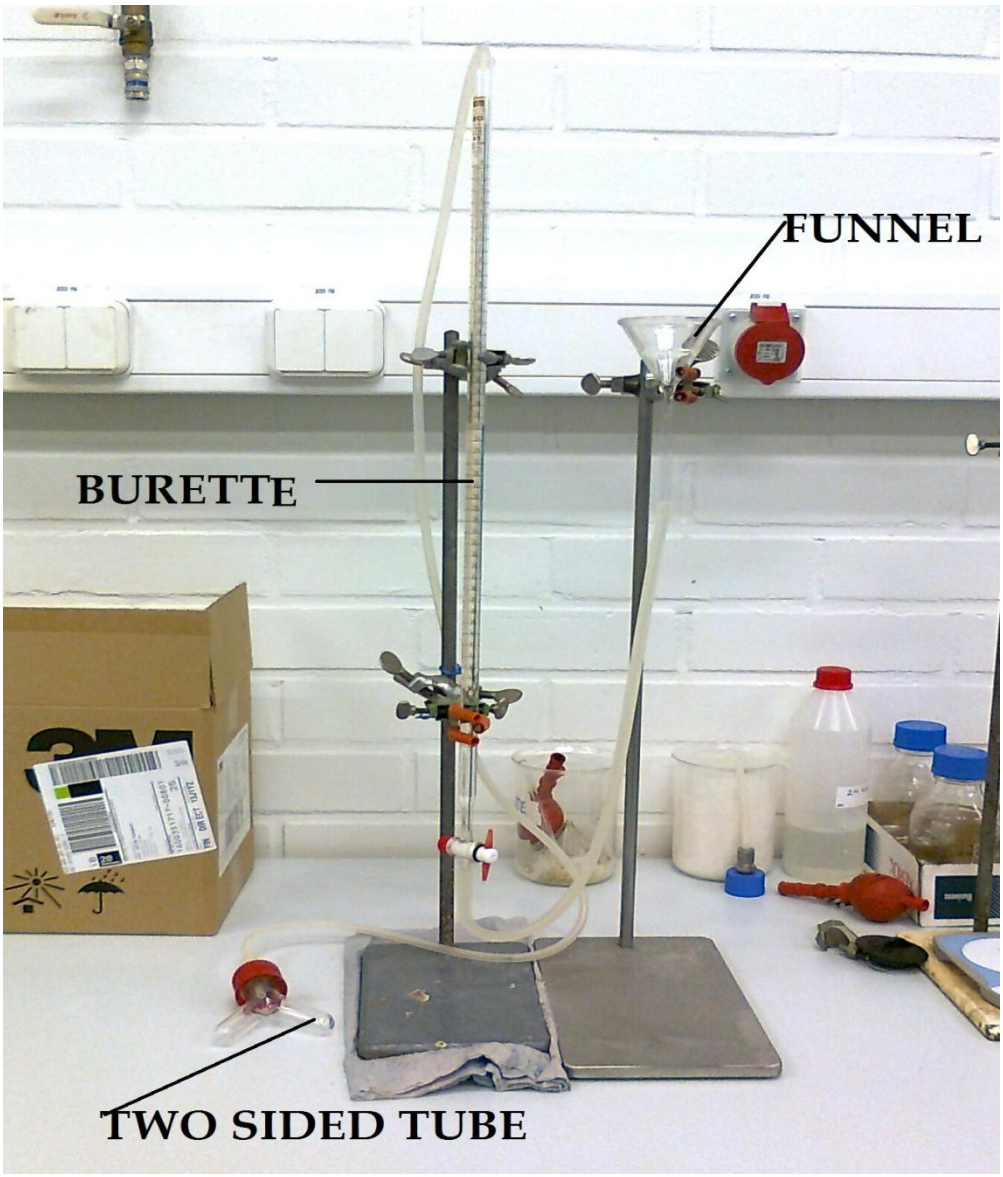
Trial Number	CO2 load (%)	Liquid flow (l/min)	Steam's pressure (bar)	Preheater's power (kW)	CO_2 removal
1	10	2.5	0.20	2	
2	10	3.0	0.25	4	
3	10	3.5	0.30	6	
4	15	2.5	0.25	6	
5	15	3.0	0.30	2	
6	15	3.5	0.20	4	
7	20	2.5	0.30	4	
8	20	3.0	0.20	6	
9	20	3.5	0.25	2	

Before the experiment plan displayed in Table 3 was executed, the absorption-desorption unit was drained of its old washing liquid and thoroughly rinsed with deionized water to remove any fouling from the packing surface. Afterwards, sev-

eral liters of 1M MEA were prepared into the two storage tanks from which they were been pumped into the absorber and the desorber continuously until the packings became adequately wet.

Having gotten the packings wet, the variables were tuned to the appropriate conditions stated in the experiment plan. Time interval of about thirty minutes was allowed in between trials to enable the system stabilize; however, the CO_2 load varies slightly in the course of the experiments. Samples of the washing liquid entering and leaving the stripper were taken for CO_2 analysis. Additionally, pressure drop and temperature measurement around the columns were also recorded for all the trials.

The CO_2 content of the amine solutions was separated and measured with the simple apparatus shown in Graph 16. As seen from Graph 16, the burette and the funnel to some extent contain the sealing liquid—a mixture of distill water, sodium sulphate salt and concentrated sulphuric acid—used to trap the released CO_2 from the two sided tube. In the glass tube on the other hand, 2ml of the MEA solution and 5ml of a 50% by weight phosphoric acid solution on the either sides were mixed for each trial's sample. The changes in the level of the burette's content were noted as the volume of CO_2 released.



GRAPH 16. CO_2 analysing equipment

The recorded data for the experimental runs and the samples analysis are presented in the next chapter.

5 DATA AND CALCULATIONS

In continuation with the CO_2 removal optimization experiment, the data collected from the experiments described in the previous chapter were utilized for some calculations. Shown below in Table 4 are the values of the CO_2 removal estimated from the collected data.

TABLE 4. CO_2 removal for the experiment trials

Trial Numbers	CO_2 removal per 2ml MEA solution (ml)		
	1st Run	2nd Run	3rd Run
1	2.35	2.3	2.25
2	1.55	3.02	2.7
3	1.15	4.05	3.75
4	7.95	5.85	7.55
5	1.9	1.35	2.95
6	2.37	2.5	2.15
7	4.95	7.55	9.3
8	4.4	7.07	5.7
9	1.35	1.9	2.1

With these data, the necessary calculations were made to determine the optimum CO_2 removal condition, the expected CO_2 removal at this condition and the contribution of each parameter to the results.

To start with, the performance parameter values (CO_2 removal) were converted to signal to noise ratio using the equation for bigger the better. The procedure is displayed below with the first trial results from Table 4.

$$S/N = -10 \times \log_{10}\left(\left({}^{1}/_{y_1^2} + {}^{1}/_{y_2^2} + {}^{1}/_{y_3^2}\right)\Big/_{n}\right)$$

$$S/N = -10 \times \log_{10}\left(\left({}^{1}/_{2.35^2} + {}^{1}/_{2.3^2} + {}^{1}/_{2.25^2}\right)\Big/_{3}\right) = 7.23$$

The same equation was used for the remaining trials to arrive at the values gathered in Table 5.

TABLE 5. Signal to noise ratios for experiment trials

Trial Number	CO2 load (%): A	Liquid flow (l/min): B	Steam's pressure (bar): C	Preheater's power (kW): D	Signal to Noise Ratio
1	1	1	1	1	7.23
2	1	2	2	2	6.56
3	1	3	3	3	5.29
4	2	1	2	3	16.81
5	2	2	3	1	5.04

6	2	3	1	2	7.33
7	3	1	3	2	16.33
8	3	2	1	3	14.67
9	3	3	2	1	4.55

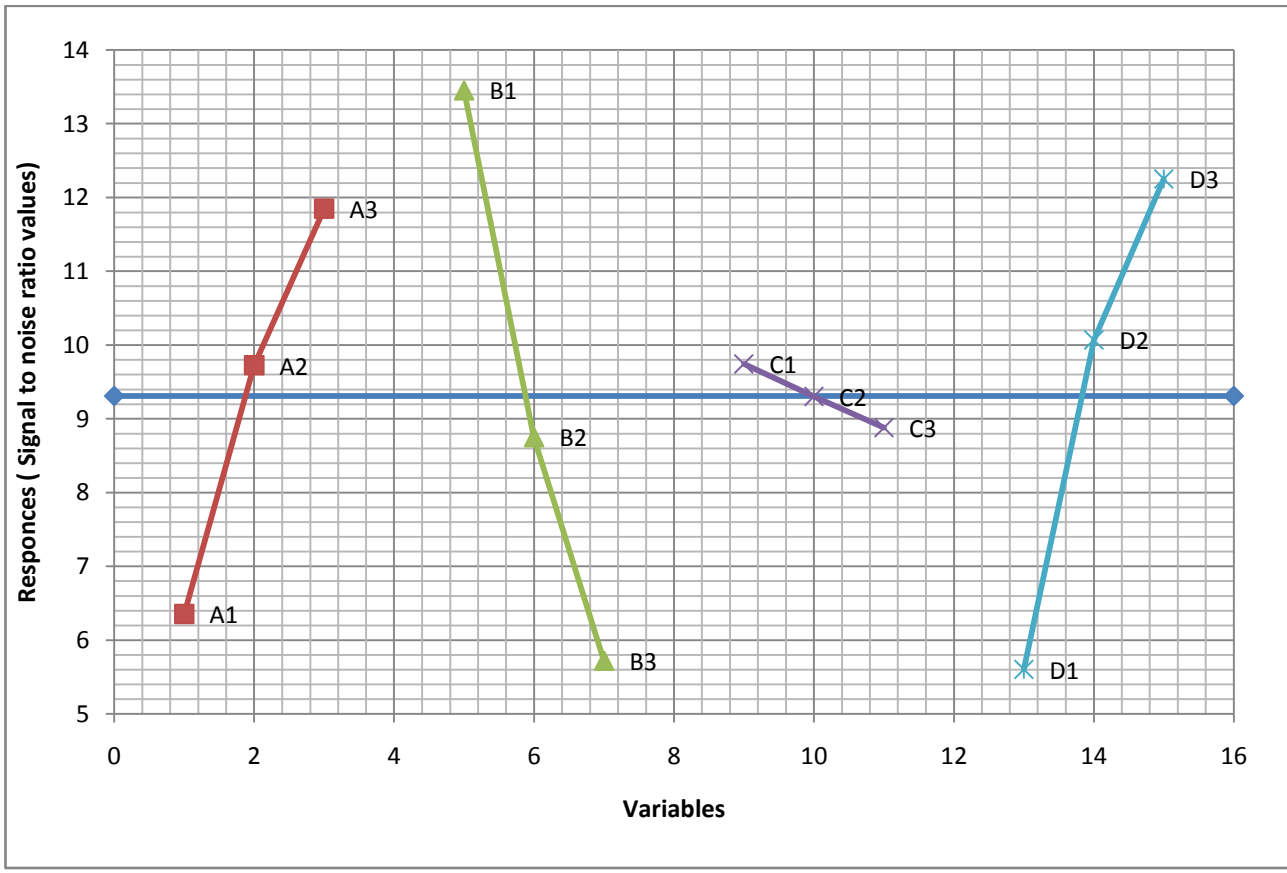
With the S/N values, the average effects of all the parameters were calculated at their stated levels. In the case of variable A (CO_2 load) ; its average performance at level1 is the average of all the S/N ratio values that include parameter A at level1, therefore:

$$A1 = {}^{7.23 + 6.56 + 5.29}/_{3} = 6.36$$

Other average effects were calculated similarly, and then used to chart the so called main effect graph from which the sought optimum condition was evident.

TABLE 6. Average effects of parameters

	Average Effects		
Variables	**Level 1**	**Level 2**	**Level 3**
CO2 load (%): A	6.357397	9.725596	11.84704
Liquid flow (l/min): B	13.45409	8.75315	5.722795
Steam's pressure (bar): C	9.743435	9.30378	8.882821
Preheater's power (kW): D	5.605564	10.07145	12.25302



GRAPH 17. Main effect plots for the four parameters

The optimum condition was chosen as the set of variables level highest on the main effect plots. This set of condition is A3 (20% CO_2 load), B1 (2.5 l/min liquid flow), C1 (0.2bar steam's pressure) and D3 (6kW preheater's power). Following this, the grand average (GA) signal to noise ratio was estimated to be 9.31.

The expected signal to noise ratio and CO_2 removal at the optimum condition were calculated with these expressions:

$$S/N_{opt} = GA + (A1 - GA) + (B3 - GA) + (C3 - GA) + (D1 - GA)$$

and $\Delta CO_{2,opt} = \sqrt{10^{(S/N_{opt})/10}}$. The expressions yielded 19.37 and 9.3ml for the expected optimum S/N ratio and the change in carbon dioxide removal respectively.

Lastly, analysis of variance (ANOVA) was performed on the data to detect the factors respective influence on the results. In the first analysis, the error factor has an infinitesimal variance. Also, it was discovered that the reboiling steam pressure influence is insignificant; thus, the analysis was repeated with factor C being pooled. The outcome of the second ANOVA calculation is shown in Table 7 below.

TABLE 7. ANOVA calculation with pooling

Column	Factors	Degree of freedom	Sum of squares	Variance	Variance ratio	Pure sum of squares	Percent contribution
1	**CO2 load (%): A**	1	45.98	45.98	206.91	45.76	22.10
2	**Liquid flow (l/min): B**	1	91.05	91.06	409.73	90.83	43.87
3	**Steam's pressure (bar): C**	pooled	pooled	pooled	pooled	pooled	pooled
4	**Preheater's power (kW): D**	1	68.89	68.89	310.00	68.67	33.17
Others/Error		5	1.11	0.22			0.86
Total		8	207.04				100

From the last column in Table 7, it can be clearly seen that variables A, B and D have significant influence on the experiment outcome. This singular reason calls for a close watch on the variation of these variables as a slight deviation from their fixed point can greatly affect the result.

In order to validate the claims made with the calculated optimum condition, three runs were made at the proposed condition. The results from these runs are presented in Table 8 below.

TABLE 8. Confirmatory tests results

Trial Condition	ΔCO2 per 2ml MEA solution (ml)		
	1st Run	2nd Run	3rd Run
A3 (20% CO_2 load), B1 (2.5 l/min liquid flow), C1 (0.2bar steam's pressure) and D3 (6kW preheater's power)	5.00	6.20	9.80

The carbon dioxide released per 2ml MEA solution from the confirmatory tests has an average of 7.00ml. This value is about 25% less than the one calculated from the stated optimum condition. A possible reason for this is the presence of interaction between some of the variables investigated or the inability of the system to stabilize as the result of constant variation of variables in the course of the trial. The trend of the figures in Table 8 also seems to suggest that the unit is not stabilized before washing liquid samples are being taken.

6 DISCUSSION

Absorption-desorption units are widely used for the purpose of carbon dioxide sequestering. The overall efficiency of these units can be tied to a number of factors amongst which is the effective washing liquid regeneration. This thesis' goals are to investigate the variables affecting CO_2 stripping, to optimize the stripping process and lastly to give recommendations on how to improve the CO_2 stripping capacity of the CO_2-MEA absorption-desorption unit whose desorber is unable to effectively regenerate its washing liquid, MEA.

In the light of these tasks, thorough research was made to elucidate the intricacies surrounding the subject matter, and also to make clear the recognition of the factors affecting CO_2 stripping. Afterwards, the Taguchi method of experimentation was employed to optimize the regeneration process.

According to literature, the following factors were found to affect the stripping ability the desorber: inlet carbon load, liquid inlet temperature; the reboiling heat, liquid flow rate; packing type, washing liquid's nature and characteristics; height of the packing, and the column diameter. The first four of the listed factors were studied in the experiment to determine their optimum combinations, as they are readily tunable. Affecting other factors suggests retrofitting the system.

The sought optimum combination of the studied variables was found to be 20% CO_2 load, 2.5 l/min washing liquid flow, 0.2bar steam's pressure and 6kW prehea-

ter's power after running twenty-seven straight experiments at nine different conditions. Theoretical estimation of carbon dioxide removal at this condition yields 9.3ml; however, the confirmatory test gave 7.0ml CO_2 removal. Carbon load, liquid flow and preheater's power were found to significantly influence the stripping process.

The notable difference between the calculated and the empirically determined CO_2 removal calls for further investigation to better achieve the optimization. This time, possible interactions within the variables should be involved in the study. Additionally, a better control system should be built around the three variables said to be substantially influencing the results.

Likewise, the capacity of the desorber can be extended by effecting the two changes discussed below:

- Usage of better packing: Raschig rings, which are currently utilized in the stripper, also in the absorber, are the oldest form of packings known. There are tenths of better random packing types with better interfacial area available for mass transfer. However, considering that the scrubber is more in size as compared to the stripper, the capacity extension that can be rendered by another random packing might be immaterial. Thus, a structural packing, known for its low pressure drop, is suggested.
- Washing solvent situation: A rough estimation of the percent CO_2 being handled in the system indicates a value above which chemically absorbing solvents are known to perform well. However, an examination of the condition of state (pressure and temperature condition) in the system favors the chemically working liquid. A change to a physically working solvent would

required higher pressure which the all-glass equipment might not be able withstand. However, this change will save cost as less heating will be required from the reboiler and at the same time better the regeneration process.

In conclusion, in order to operate this equipment with better washing liquid regeneration, the four-variable optimizing condition stated above must be adhered to. However, improving the capacity of the desorber involves the retrofitting actions stated above as well.

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APPENDIX 1/1

Raw data collected from the experiments were presented below.

First run

Experi-peri-ment Num-ber	Tgas, in deg, C	Tgas, out deg, C	Tliq, in deg, C	Tliq, out deg, C	Tliq, in(Strip per) deg, C	Strip per PD mbar	Ab-sorber PD mbar	CO2 load in %	CO2 load out %	CO2 in ml	CO2 out ml
1	16.0	12.6	10.7	9.5	55.1	1.8	3.6	10.05	6.70	20	17.65
2	16.7	10.0	10.0	10.1	64.9	2.2	4.0	10.18	7.3	19.85	18.3
3	16.5	10.4	11.0	11.2	71.9	2.7	4.3	9.48	7.53	19.6	18.45
4	16.6	12.2	9.6	11.1	85.7	50.2	4.9	14.9	9.12	20.05	12.1
5	16.7	10.8	9.8	11.6	54.9	2.7	5.4	14.26	10.95	21.0	19.1
6	14.7	10.9	10.9	11.0	62.5	3.1	7.2	15.60	11.97	22.9	20.53
7	7.9	9.2	10.5	11.8	70.7	3.2	5.2	20.66	9.06	23.65	18.7
8	11.8	10.2	12.1	13.9	80.3		5.9	19.66	12.62	22.05	17.65
9	11.9	10.6	13.2	13.7	54.7		7.8	19.80	14.32	23.35	22.0

APPENDIX 1/2

Second run

Experi-peri-ment Num-ber	Tgas, in deg. C	Tgas, out deg. C	Tliq, in deg. C	Tliq, out deg. C	Tliq, in(Stripper) deg. C	Stripper PD mbar	Ab-sorber PD mbar	CO2 load in %	CO2 load out %	CO2 in ml	CO2 out ml
1	17.9	9.8	12.0	11.2	56.1	1.1	4.0	10.63	6.17	24.5	22.2
2	18.8	10.4	13.4	12.7	68.1	1.4	5.35	9.8	7.31	24.75	21.73
3	19.3	10.8	13.4	13.9	75.2	55	6.1	10.7	7.95	24	19.95
4	16.6	9.8	10.8	12.8	85.9	46	5.4	16.0	9.71	23.35	17.5
5	15.8	10.5	12.2	12.8	59.4	-2.4	6.4	15.44	11.32	22.9	21.55
6	16.6	11.3	13.6	13.9	64.2	-0.2	8.0	15.22	12.22	25.95	23.45
7	12.5	9.3	11.8	13.5	74.9	52.6	6.5	20.50	14.59	25.75	18.2
8	13.3	10.7	14.5	13.8	81.4	20	8.2	19.56	15.64	25.3	18.23
9	14.5	10.5	13.9	13.8	56.1	-2.7	9.0	19.6	16.16	25.45	23.55

APPENDIX 1/3

Third run

Experi-peri-ment Num-ber	Tgas, in deg. C	Tgas, out deg. C	Tliq, in deg. C	Tliq, out deg. C	Tliq, in(Stripper) deg, C	Stripper PD mbar	Absorber PD mbar	CO2 load in %	CO2 load out %	CO2 in ml	CO2 out ml
1	15.5	8.7	12.0	11.9	45.31	2.1	3.9	10.0	6.04	25.5	23.25
2	15.9	8.6	12.9	13.3	67.8	1.4	4.9	10.2	7.03	25.45	22.75
3	17.7	10.2	12.9	14.5	76.0	58.9	8.8	10.15	8.79	25.25	21.5
4	15.1	9.2	11.2	13.2	86.4	50.2	6.1	15.04	10.64	25.9	18.35
5	12.9	10.1	13.8	12.8	55.7	1.8	5.9	15.50	9.81	27.95	25.0
6	13.8	11.2	14.8	14.2	62.2	1.6	7.4	15.20	11.35	28.35	26.2
7	10.9	9.7	12.2	13.1	70.2	55.2	7.3	20.20	15.15	30.05	20.75
8	10.8	10.3	13.8	14.7	79.2	57.7	8.2	20.05	16.04	28.2	22.5
9	9.7	10.9	15.4	14.5	51.7	1.2	9.3	20.50	15.06	29.9	27.8

APPENDIX 2/1

The first version of the ANOVA calculation is shown below.

COLUMN	FAC-TORS	DEGREE OF FREE-DOM	SUM OF SQUARES	VA-RIANCE	VA-RIANCE RATIO	PERCENT CONTRIBU-TION
1	**CO2 load (%): A**	1	45.9815183	45.981518	--	22.20904
2	**Liquid flow (l/min): B**	1	91.0548369	91.054837	--	43.97942
3	**Steam's pressure (bar): C**	1	1.1111579	1.1111579		0.536688
4	**Prehea-ter's power (kW): D**	1	68.892129	68.892129	--	33.27485
Oth-ers/Error		4	4.5475E-13	--	--	--
Total		8	207.039642			100

APPENDIX 3/1

The general flow diagram of the CO2-MEA absorption-desorption unit is depicted below.