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Performance evaluation and optimisation of post combustion CO_2 capture processes for natural gas applications at pilot scale via a verified rate-based model

Fatemeh Rezazadeh[a], William F. Gale[a], Muhammad Akram[b], Kevin J. Hughes[b], Mohamed Pourkashanian[b]

[a]Centre for Integrated Energy Research (CIER), School of Chemical and Process Engineering, The University of Leeds, Leeds, LS2 9JT, UK

[b]Department of Mechanical Engineering, University of Sheffield, Sheffield, S1 3JD, UK

Abstract

CO_2 absorption based on chemical reactions is one of the most promising technologies for post combustion CO_2 capture (PCC). There have been significant efforts to develop energy efficient and cost effective PCC processes. Given that PCC is still maturing as a technology, there will be a continuing need for pilot scale facilities to support process optimisation, especially in terms of energy efficiency. Pilot scale PCC facilities, which are usually orders of magnitude smaller than those that will be used in future in large scale fossil power plants, make it possible to study details of the PCC process at an affordable scale. However, it is essential that pilot scale studies provide credible data, if this is to be used with confidence to envisage the future large-scale use of the PCC process, especially in terms of energy consumption. The present work therefore establishes and experimentally verifies (using a representative pilot plant as a case study) procedures for analysing the energy performance of a pilot scale amine based CO_2 capture plants, focusing on natural gas fired applications. The research critically assesses the pilot plant's current energy performance, and proposes new operating conditions and system modifications by which the pilot plant will operate more efficiently in terms of energy consumption. The methodology developed to assess and improve the energy performance of the PCC process is applicable, with appropriate inputs, to other plants of this type that employs aqueous 30 wt. % monoethanolamine (MEA) solution as the solvent. A rate based model of the post combustion CO_2 capture process using an aqueous solution of 30 wt. % MEA as the solvent was developed in Aspen Plus® V.8.4, and verified using the results of experimental studies carried out using the UK Carbon Capture and Storage Research Centre / Pilot-scale Advanced Capture

Technology (UKCCSRC/PACT) pilot plant, as a representative pilot-scale capture plant, and employed for parametric sensitivity studies. Several parameters have been identified and varied over a given range of lean solvent CO_2 loading to evaluate their effects on the pilot plant energy requirement. The optimum lean solvent CO_2 loading was determined using the total equivalent work concept. Results show, for a given packing material type, the majority of energy savings can be realised by optimising the stripper operating pressure. To some extent, a higher solvent temperature at the stripper inlet has the potential to reduce the regeneration energy requirement. A more efficient packing material, can greatly improve the pilot plant overall energy and mass transfer efficiency.

Key words: Post-combustion CO_2 capture, energy consumption, specific energy requirement, total equivalent work, MEA,

1. Introduction

A post combustion CO_2 capture (PCC) process based on chemical absorption using aqueous solutions of amine as solvent is the most mature CO_2 capture technology, with the 30 wt. % aqueous solution of monoethanolamine (MEA) as the base-line solvent (1). Despite this process having been used for many years in various industrial applications, such as natural gas treatment plants (2,3), there are considerable challenges in its utilisation to partially decarbonise fossil fuel power plants. The largest existing industrial absorption plants are orders of magnitude smaller than those that would be installed in a medium to large-scale power plant. For instance, major equipment such as the absorber tower and stripper column required to serve a large-scale power plant are larger than any of their kind that have been built before (3). To successfully employ this technology in large-scale plant, detailed scaled up based on pilot studies and optimisation studies, based on reliable and predictive simulation models are necessary. Furthermore, future advancements of this technology, after the initial implementation, will need to be tested via pilot scale studies prior their use. If scale up is to be achieved, it is essential that data from the pilot plant is both credible and applicable. The present work therefore establishes and experimentally verifies (using a representative pilot plant) procedures for analysing the initial set-up and operation of pilot scale amine based CO_2 capture plants. The authors have chosen to focus on natural gas fired plant, given that natural gas is a relatively clean fuel, compared with coal and hence may have a longer term future, but is not a truly low carbon source of electric power and hence has a need for carbon capture.

A crucial challenge associated with the MEA-based CO_2 capture process is its large energy requirement, especially for the solvent regeneration which takes place in the stripper. Studies have shown that the addition of an amine-based CO_2 capture unit to a natural gas combined cycle power plant leads to a net power plant efficiency penalty of 7-11 % (4,5).

More than 70 % of the total energy a CO_2 capture process requires is used for the solvent regeneration (8). As reported in the literature, the specific regeneration energy requirement of a CO_2 capture process using 30 wt.% MEA as solvent to remove 90 % CO_2 of natural gas fired flue gases seems to converge to values of around 3.2 to 4.2 MJ per kg of CO_2 captured (3,6,7). Therefore, reducing the regeneration energy requirement has globally been the focus of many research and development (R&D) studies such as CASTOR (9), CESAR (10), etc. In addition to developing new solvents with better overall performance than MEA, many research studies have investigated the benefits of modifying the conventional CO_2 capture process or identifying ideal operating conditions to optimise its performance in terms of energy consumption (7,11-18). Some of these studies have resulted in setting up pilot plants (15,18-20) to ascertain claimed benefits of proposed scenarios. In the majority of the studies that have been reported, aqueous solutions of MEA were usually taken as the base-line solvent, to which new solvents were compared.

Process modelling is usually required for a better understanding of chemical processes, evaluating alternate process configurations before their experimental assessment, and troubleshooting of the process in case of malfunction. In addition, to design and scale-up a pilot-scale CO_2 capture process to a capacity suitable for commercial scale power plant applications, reliable process modelling is essential. To achieve this, models need to reliably represent the physical and chemical equilibria in the system and also accurately account for mass transfer and reaction kinetics. Such models are developed based on information of physical and chemical properties of the reactive components and validated using pilot plant data (21). To model a chemical absorption process, for which the amine-based CO_2 capture is an example, rate-based modelling is the most reliable method. Equilibrium stage models, despite often being suitably applied to describe distillation and reactive distillation processes (22-24-14), usually fail to adequately simulate a reactive absorption process (22,25,26).

This study aims to assess operating conditions and energy consumption of a typical PCC process for natural gas fired applications using 30 wt. % MEA as solvent via modelling and accordingly propose process modifications and operating conditions, suitable for testing in pilot plants, by which the process operates more efficiently in terms of energy consumption. A rate-based model of the CO_2 capture process was developed in Aspen Plus® V.8.4, and verified using results of experimental studies carried out using the UK Carbon Capture and Storage Research Centre / Pilot-scale Advanced Capture Technology (UKCCSRC/PACT) pilot plant, denoted as the PACT pilot plant in this paper for simplicity. The PACT pilot plant was considered as a representative PCC process and a number of parametric studies were carried out to determine its optimal operating conditions. Results of the CO_2 capture model verification and discussions on the proposed process modifications and operating conditions are presented in this paper.

2. Case study pilot plant and process description

2.1. Process description

The design of the PACT pilot plant is based on a standard amine-based CO_2 capture plant. Figure 1 schematically shoes the gas turbine arrangement and its connection with the PACT amine CO2 capure pilot plant. The one tonne per day CO_2 capture plant uses 30 wt. % MEA as solvent and operates with the flue gas provided by a 100 kW_e micro gas turbine (Turbec T100). The micro gas turbine, which is a combined heat and power unit, consists of a centrifugal compressor, radial turbine and high speed generator, which all are mounted on one shaft (27). Natural gas burns in the combustor and the hot flue gas expands through the turbine diffuser with an average CO_2 concentration of 1.6 % (on a molar basis; all subsequent CO_2 concentration percentages are on a molar basis unless otherwise state). To attain a flue gas with conditions similar to that of a natural gas fired combined cycle power plant, i.e. 4 to 6 % CO_2 concentration, the turbine flue gas was mixed with CO_2 gas from a CO_2 storage tank. The flue gas CO_2 concentration was then increased in four steps up to 9.9 % to resemble flue gas conditions similar to a gas turbine with an exhaust gas recirculation (EGR) cycle at various recycle rates. The experiments presented in this study were carried out by injecting only pure CO_2 gas to the flue gas stream without adding any other traces such as NO_x or SO_2.

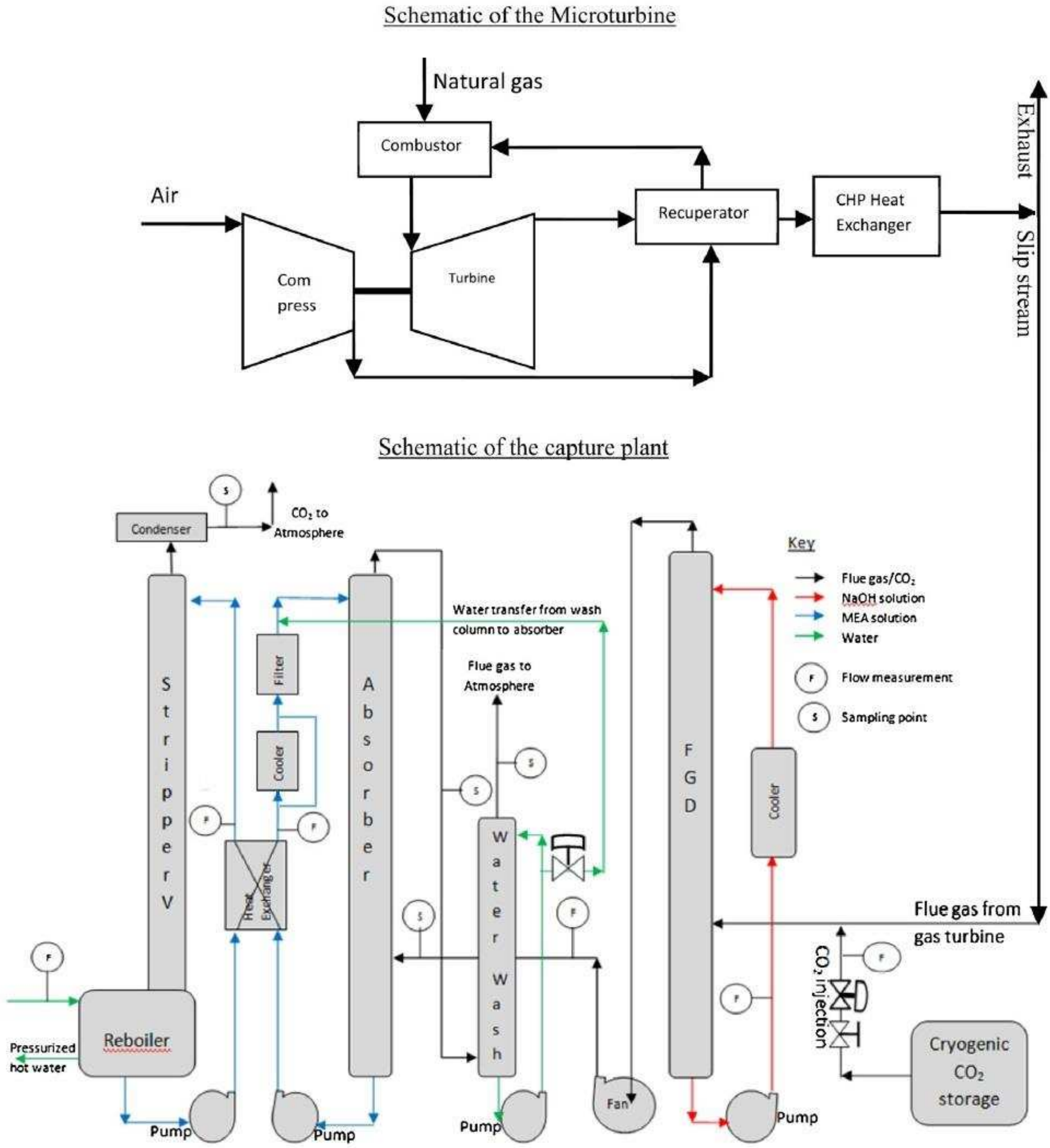

Figure 1. Schematic overview of the UKCCSRC/ PACT micro gas turbine, amine CO_2 capture plant and their integration with CO_2 injection system (28)

The pressure of the flue gas is increased by a booster fan before entering the absorber column. The typical 40 °C flue gas temperature at the absorber inlet was achieved by controlling the gas turbine heat exchanger bypass flow rate. An orifice plate flow meter along with temperature and pressure indicators measures the flue gas conditions at the absorber inlet. The flue gas flow rate throughout the experiments was constant due to plant operating conditions. However, the solvent flow rate was varied with the variation of the flue gas CO_2 partial pressure to maintain a fixed CO_2 removal rate.

The pilot plant consists of a packed absorber column, a packed water-wash column, and a packed stripper column constructed in a similar fashion to the absorber column with an air-cooled condenser and a reflux drum at the top. Columns are packed with INTALOX Metal Tower Packing (IMTP) No. 25 random packing due to its low cost and ease of installation. Table 1 summarises the pilot plant design specifications. Heat integration of the regenerated and rich solvent is realised via a plate type heat exchanger, and further cooling of the lean solvent prior entering the absorber column is achieved by an air-cooled induced draft cooler.

Table 1. The UKCCSRC/PACT design specifications

Parameter	Specification
Flue gas source	Turbec T100 micro gas turbine + CO_2 feed from CO_2 storage tank
CO_2 concentration in the flue gas	5.5-9.9 %
Flue gas flow rate in the absorber	250 Nm^3/h
Flue gas temperature at the absorber inlet	~ 40 °C
Solvent type	30 wt. % MEA aqueous solution
Solvent flow rate	~ 400-1200 kg/h
Solvent temperature at the absorber inlet	40 °C
Column packing in absorber, stripper, water washing sections	Koch IMTP25 random packing
Material of packing	metal
Diameter of columns (absorber, stripper, water wash sections)	0.30 m
Height of packing	
Absorber	8 m
Stripper	8 m
Water wash	1.2 m
Pressure in the absorber	Atmospheric pressure
Pressure in the stripper	120 – 300 kPa absolute

The counter-current contact of the flue gas entering the absorber column below the packing section with the lean solvent solution entering above the packing section results in the absorption of CO_2 by the solvent. Before the treated gas leaves the absorber column, it has to pass a demister to retain carried over liquid droplets. To further reduce amine losses, the flue gas leaving the absorber enters the wash column where it is treated with water to remove droplets of amine before exiting to atmosphere.

The temperature and mass flow rate of the lean solvent entering the absorber column are controlled. A Coriolis flow measurement device measures the lean solvent flow rate, and the required flow rate is controlled by a proportional control valve. The lean solvent temperature is measured by a thermocouple at the absorber inlet and controlled by opening of the valve bypassing the lean solvent across the lean solvent air-cooler. A Coriolis flow measurement device measures the rich solvent flow rate leaving the absorber column. The composition of the rich solvent can be determined by analysis of a liquid sample taken downstream of the rich solvent pump. To ensure the plant steady state operation, the rich solvent level in the absorber sump is controlled by the rich amine pump.

Before being fed to the stripper column, the rich solvent is pumped through the cross heat exchanger to be heated up by the hot lean solvent leaving the stripper column, and both stream temperatures at the heat exchanger inlet and outlet are measured. The rich solvent enters the stripper column above the packed section, and the product vapour leaves the stripper from the top. The stripping steam is generated at the stripper bottom by partial evaporation of the liquid solvent in the reboiler, with the heat required in the reboiler being provided by pressurised hot water. The mass flow rate, inlet and outlet temperatures of the hot water are measured and recorded to calculate the heat required for solvent regeneration. The hot lean solvent leaves the stripper from the bottom and flows through the cross heat exchanger and the air-cooler to enter the absorber column. The composition of the lean solvent can be determined by analysis of a liquid sample taken downstream of the lean solvent pump.

To obtain temperature profiles for the absorber column, temperature was measured along the whole length of absorber column at different locations of 2m, 3.3m, 5.1m, and 6.8m in height from the gas entry point. Along the stripper, temperature was recorded at 0.3m (bottom), 3.8m (middle) and 7.5m (top) heights from the bottom of the stripper.

Two Servomex analyzers – a Servomex 4900 for O_2 and low level CO_2 measurement, as well as a Servomex 2500 for high level CO_2 measurement were used to analyse the flue gas composition at the following locations: inlet of the absorber, exit of the absorber, exit of the wash column and CO_2

concentration at the exit of the stripper. The Servomex 4900 draws samples from three locations (absorber inlet, absorber outlet, wash column outlet) alternately. The switchover happens every 5 minutes and is controlled by a programmable logic controller (PLC) through solenoid valves. In order to avoid condensation problems, the temperature of the heated sampling lines was maintained at 150 °C in all cases. The sampling points have been equipped with coalescence filters to remove droplets of water carried over by the gas. The alkalinity of the solvent is determined analytically by titrating samples with HCl solution, while the CO_2 loading of the lean and rich solutions are determined via titrating samples with NaOH solution. The control of the pilot plant is done via programmable logic controllers (PLCs) while data acquisition and logging are performed with LABVIEW® interfaced with MS Excel®.

2.2. Experimental data

As mentioned earlier, for these experimental tests, the CO_2 concentration in the flue gas at the absorber inlet was varied in steps from 5.5 % to 9.9 %. The plant is capable of treating flue gas flow rates up to 250 Nm^3/h. For these tests, the flue gas flow rate was maintained at around 210 Nm^3/h and its temperature was controlled at 40 °C. The solvent flow rate was varied to change the L/G ratio corresponding to different CO_2 concentrations to maintain a constant CO_2 capture rate of 90 %. An aqueous solution of nearly 30 wt. % MEA was used as the solvent, and the temperature of lean solvent at the absorber inlet was controlled at 40 °C. The 30 wt. % MEA was chosen as this is the baseline concentration used widely in absorption based CO_2 capture studies (1,3,4,7). In addition, higher concentrations of MEA solution are known to cause corrosion problems and elevate the risk of solvent carry over to the atmosphere (29). The control mechanism of the plant kept the lean solvent flow constant in order to fix the liquid to gas ratio (L/G) in the absorber, for a particular test. However, the rich solvent flow rate was varied in order to control the levels in the stripper and the absorber. Hot pressurised water at pressure of 400 kPa and temperature not higher than 120 °C was used as the reboiler heat source, and its flow rate was controlled at 7.43 m^3/h. Table 2 summarises the key process characteristics of these experimental tests.

Table 2. Process characteristics of test campaigns with variable flue gas CO_2 concentration (18)

Parameters	Unit	Case#1	Case#2	Case#3	Case#4	Case#5
CO_2 in flue gas (after CO_2 injection)	vol. %	5.5	6.6	7.7	8.3	9.9
CO_2 mass flow rate in flue gas (after injection)	kg/h	21.1	25.4	29.6	31.9	38
Solvent flow	kg/h	400	488	567	604	721
Hot pressurised water (HPW) flow	m^3/h	7.43	7.43	7.43	7.43	7.43
HPW temperature at reboiler inlet	°C	120.6	120.4	120.8	120.5	120.5
HPW temperature at reboiler outlet	°C	115.8	114.5	115.3	114.5	114.7
Cold approach temperature $(T_{ap})_C$	°C	19.03	18.44	19	18.50	19.8
Hot approach temperature $(T_{ap})_H$	°C	19.72	18.99	20.03	19.84	19.17
Rich solvent concentration	wt. %	30.8	27.8	30.6	27.5	29.1
Lean solvent concentration	wt. %	31.9	29.9	31.7	29.8	30.5
Rich loading	mol CO_2/mol MEA	0.388	0.399	0.411	0.417	0.443
Lean loading	mol CO_2/mol MEA	0.165	0.172	0.183	0.18	0.204
Degree of regeneration	%	57.5	56.9	55.5	56.8	54.0
Liquid to Gas ratio	kg/kg	1.55	1.88	2.17	2.30	2.73
Solvent to CO_2 ratio	kg/kg	19.9	20.6	21.1	20.7	21.7
Specific Reboiler duty	MJ/kg CO_2	7.1	7.4	6.0	6.1	5.3
Stripper bottom temperature	°C	110.4	108.8	109.7	108.8	108.8
Stripped CO_2 mass flow rate	kg/h	19.5	23.2	26.7	28.9	34.3
CO_2 removal rate	%	90.8	90.3	90.0	90.2	90.8

3. Methodology

3.1. Simulation

The CO_2 absorption/desorption process with 30 wt.% MEA solution was modelled using the RateSep^{TM} model, a rigorous framework to model rate-based separations in Aspen Plus$^{®}$ V.8.4. The model used for the thermodynamic properties is based on the work done by Zhang et al. (30) who validated it against experimental data available in literature. The model uses the asymmetric electrolyte non-random-two-liquid (e-NRTL) property method to describe the liquid phase activity coefficients, and the Redlich-Kwong (RK) equation of state for the vapour phase properties (31). The absorber model comprises both equilibrium and kinetic rate-based controlled reactions, while the stripper model comprises equilibrium rate-based controlled reactions, and the reboiler section in the stripper column was modelled as an equilibrium stage. In this study, packed columns were divided into 20 identical segments (stages). In the absorber column, the reactions that involve CO_2 were described with a kinetic model. The equilibrium reactions describing the solution chemistry of CO_2 absorption with MEA, which are integral components of the thermodynamic model, are expressed as (30):

$$2H_2O \leftrightarrow H_3O^+ + OH^- \quad (1)$$

$$CO_2 + 2H_2O \leftrightarrow HCO_3^- + H_3O^+ \quad (2)$$

$$HCO_3^- + H_2O \leftrightarrow H_3O^+ + CO_3^{2-} \quad (3)$$

$$MEAH^+ + H_2O \leftrightarrow MEA + H_3O^+ \quad (4)$$

$$MEACOO^- + H_2O \leftrightarrow MEA + HCO_3^- \quad (5)$$

199 The following describes the forward and reverse reactions of bicarbonate and carbamate formation,
200 respectively (32):

$$CO_2 + OH^- \rightarrow HCO_3^- \quad (7)$$

$$HCO_3^- \rightarrow CO_2 + OH^- \quad (8)$$

$$MEA + CO_2 + H_2O \rightarrow MEACOO^- + H_3O^+ \quad (9)$$

$$MEACOO^- + H_3O^+ \rightarrow MEA + CO_2 + H_2O \quad (10)$$

201 The Aspen RateSep™ model requires quantitative values of transport properties that are essential for
202 correlations of heat transfer, mass transfer, interfacial area, liquid holdup, pressure drop, etc.
203 (30,32,33). The transport properties include density, viscosity, surface tension, thermal conductivity,
204 and binary diffusivity (33). Table 3 summarises the models with their literature references used in
205 Aspen Plus for transport property calculations.

206 Table 3. Transport property models used in Aspen Plus for the CO_2 capture model (30,32,33)

Property	Model used
Mass transfer at vapour-liquid interface	Two-film theory
Thermo-physical property model	Ying and Chen model
Liquid density	Clarke density model
Gas density	Redlich-Kwong equation of state
Liquid viscosity	Jones-Dole electrolyte correction model
Gas Viscosity	Chapman−Enskog model with Wilke approximation
Thermal conductivity of the liquid	Riedel electrolyte correction model
Surface tension of the liquid solution	Onsager-Samaras model
Diffusivity of CO_2 in H_2O and MEA-H_2O solutions	Wilke-Chang diffusivity model

207 ### 3.2. Process Evaluation

208 To evaluate the energy performance of the PACT pilot plant, the total equivalent work concept is used
209 in addition to the specific regeneration energy requirement. This concept estimates the total electrical
210 work penalty that would be imposed on the power plant by operating the CO_2 capture plant. Eq. 1
211 shows the three main contributors to the total equivalent work (11):

$$W_{eq} = W_{heat} + W_{comp} + W_{pump} \quad (1)$$

Where, W_{eq} is the total equivalent work, W_{heat} is the regeneration heat equivalent work, W_{comp} is the compression equivalent work and W_{pump} is the pump equivalent work. The equivalent electrical penalty associated with solvent regeneration, called the regeneration heat equivalent work, is calculated using the Carnot efficiency method, as represented by Eq. 2 (11):

$$W_{heat} = \eta_{turbine}\left(\frac{T_{reb} + \Delta T - T_{sink}}{T_{reb} + \Delta T}\right) Q_{reb} \quad (2)$$

Where, $\eta_{turbine}$ is the Carnot efficiency, T_{reb} is the solvent temperature at the reboiler, ΔT is the temperature difference between hot and cold streams at the reboiler, T_{sink} is the cooling water temperature, and Q_{reb} is the reboiler heat duty. Assumptions made for Eq. 2 include a 90 % efficiency to account for non-ideal expansion in steam turbines (34), an approach temperature of 5 °C for the steam side in the reboiler section, and a sink temperature of 40 °C.

The compression work is the work required to compress the captured CO_2 from the stripper pressure (P_{in}), to the storage pressure, e.g. 15 MPa (150 bar), and calculated using Eq. 3 (35).

$$W_{comp} = -3.48 \ln(P_{in}) + 14.85, \qquad 1 < P_{in}\,(bar) < 20 \quad (3)$$

Assumptions made for Eq. (3) include a compression ratio of 2 or less for each compression stage, a compressor polytropic efficiency of 86 %, inter-stage cooling to 40 °C with knocked out water between stages with zero pressure drop (35).

The pump work includes only the required head at the efficiency of the pump, e.g. 75 %, to move and circulate the solvent from the absorber to the pressure of the stripper and vice versa. The flue gas blower work is excluded from this calculation, assuming the flue gas pressure at the absorber inlet is sufficiently high to overcome the passage and packing pressure drops. The Aspen Plus pump block is used to calculate the pump work.

4. Results and discussion

4.1. Model verification

Experimental data presented in Table 2 were used to verify the accuracy and reliability of the developed rate-based model. The verification results were presented in Tables 4 and 6.

Table 4. Comparison of experimental and simulation results of operating parameters

Description		Rich solvent CO_2 loading (mol /mol)	Solvent temperature at reboiler (°C)	Captured CO_2 mass flow rate (kg/h)	Reboiler heat duty (kW)
Case#1	Experiment	0.388	110.4	20.2	40.0
(5.5 % CO_2)	Simulation	0.394	110.0	20.3	41.77
Case#2	Experiment	0.399	108.8	23.76	48.6
(6.6 % CO_2)	Simulation	0.411	108.5	24.3	45.6
Case#3	Experiment	0.411	109.7	26.9	45.0
(7.7 % CO_2)	Simulation	0.414	109.8	28.7	48.9
Case#4	Experiment	0.417	108.8	29.2	49.4
(8.3 % CO_2)	Simulation	0.426	108.8	30.6	49.6
Case#5	Experiment	0.443	108.8	33.2	48.5
(9.9 % CO_2)	Simulation	0.443	108.8	36.1	50.3

The absolute deviation of a simulated result from the experimental one was calculated using Eq. (4):

$$\text{Deviation (\%)} = \frac{\left|i_{experiment} - i_{simulation}\right|}{i_{experiment}} \times 100 \quad (4)$$

The mean absolute deviation values of the parameters compared in Table 4 are in the range of 0.15 to 4.7 percentages which are within an acceptable range.

To characterise the process independent of scale, performance parameters as defined in Table 5 were used.

Table 5. Parameters to characterise the plant performance independent of the scale

Parameter	Definition
CO_2 removal rate	$\psi_{CO_2} = m_{CO_2}^{TG} / m_{CO_2}^{FG}$
Degree of regeneration (mol/mol)	$\Delta x_{reg} = \left(x_{CO_2}^{rich} - x_{CO_2}^{lean}\right) / x_{CO_2}^{lean}$
Specific regeneration energy requirement (MJ/kg CO_2)	$Q_{specific} = Q_{reboiler} / m_{CO_2}$
Absorption capacity (kg/kg)	$C_{abs} = m_{CO_2} / m_L$

Where, $m_{CO_2}^{TG}$ is CO_2 mass fraction in the treated gas at the absorber outlet, $m_{CO_2}^{FG}$ is CO_2 mass fraction in the flue gas at the absorber inlet, $x_{CO_2}^{rich}$ is the rich solvent CO_2 loading. $x_{CO_2}^{lean}$ is the lean solvent CO_2 loading, $Q_{reboiler}$ is the reboiler heat duty, m_{CO_2} is the mass flow rate of CO_2 captured, and m_L is the mass flow rate of lean solvent.

Table 6. Comparison of experimental and simulation results of performance parameters

Case		Ψ_{CO_2} (%)	Δx_{reg} (%)	$Q_{specific}$ (kJ/kg CO_2)	C_{abs} (g/kg)
Case#1	Experiment	90.8	57.5	7.1	50.3
(5.5 % CO_2)	Simulation	94.9	58.1	7.3	50.7
Case#2	Experiment	90.3	56.9	7.4	48.6
(6.6 % CO_2)	Simulation	94.7	58.2	6.8	49.8
Case#3	Experiment	90.0	55.5	6.0	47.5
(7.7 % CO_2)	Simulation	96.0	55.8	6.1	50.6
Case#4	Experiment	90.2	56.8	6.10	48.2
(8.3 % CO_2)	Simulation	95.0	57.7	5.8	50.6
Case#5	Experiment	90.8	54.0	5.30	46.1
(9.9 % CO_2)	Simulation	94.1	54	5.0	50.1

The mean absolute deviation values of the parameters compared in Table 6 are in the range of 1.1 to 5.0 percentages which are also within an acceptable range.

Figure 2 shows the absorber temperature profile along the column height. The temperature was measured at 2, 3.3, 5.1 and 6.8 m heights from the gas entry point at the bottom of the column. The temperatures shown in the Figure at 0 m and 8 m heights are measured in the gas stream, not inside the absorber and are that of the flue gas entering the absorber column and leaving the absorber column. Hence, to plot the simulated temperature profiles, the flue gas inlet and outlet temperatures, that are inputs of the simulations, were used for these two points.

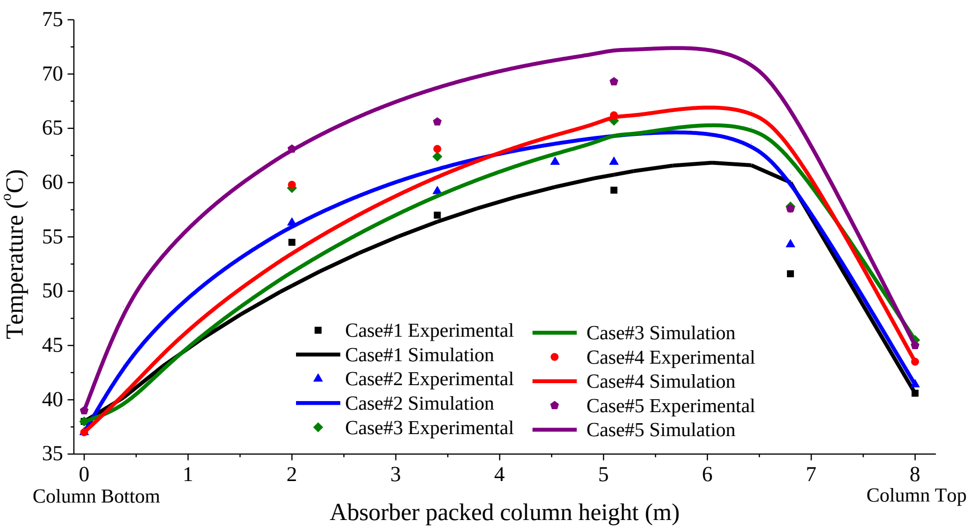

Figure 2. Absorber temperature profile based on experimental results vs. simulation results for 5 study cases (the temperatures at 0 m and 8 m are that of the flue gas at inlet and outlet of the absorber column, respectively)

As presented in Table 6, simulated CO_2 removal rates were on average converged to nearly 95 % in all five cases whilst those of the experiments were around 90 %. The constant difference of nearly 5 % between the simulated and experimental CO_2 removal rate indicates that the mass transfer efficiency in the absorber column is sub-optimal, and points out the possibility of poor solvent distribution over the absorber packed column. Furthermore, the specific regeneration energy requirement corresponding to each experiment is sub-optimal and considerably higher than what has been reported to be attainable in industry to date, i.e. (3.2-4.2 MJ per kg of CO_2 captured using 30 wt.% MEA solvent (5)). These two issues underscore the need for some modelling work to be carried out to identify the appropriate system modifications and operating conditions by which the pilot plant operates optimally for a given flue gas condition. As the results of the developed model showed good agreement with the experimental data, it is therefore meaningful to employ the model for further studies. This also illustrates that modelling and experimental activities can complement each other, and both should possibly run concurrently to deliver reasonable results.

4.2. Energy analysis

Having validated the developed rate-based model using the PACT pilot plant experimental results over a range of flue gas conditions, application of the model to improve plant design was then demonstrated, using the PACT pilot plant as a case study, specifically the test case with 5.5 % CO_2 flue gas (case#1). The 90 % CO_2 removal rate was targeted using the flue gas condition presented in Table 7.

Table 7. The base-case performance characteristics

Parameter	Value
Total flue gas mass flow rate	260 kg/h
Flue gas temperature at absorber inlet	40 °C
Flue gas pressure at absorber inlet	~ 125 kPa
Flue gas composition	
N_2	74.74 %
O_2	16.6 %
CO_2	5.5 %
H_2O	3.16 %

Specific regeneration energy and total equivalent work were chosen as parameters independent of scale to evaluate and compare the process energy performance. Four areas of improvement were identified to be explored which have potential applicability to improve the performance of a CO_2 capture process: solvent lean loading, cross heat exchanger logarithmic mean temperature difference (LMTD), stripper operating pressure, and replacement of the current packing, i.e. IMTP25, with a more efficient packing, i.e. Sulzer Mellapak 250Y.

4.2.1. Solvent lean loading

The stripper energy consumption is strongly dependant on the lean solvent CO_2 loading. For a given rich loading, if lean loading increases, the amount of steam required per unit of produced CO_2 will be reduced. Increasing lean loading can be achieved by increasing solvent circulating rate with respect to the targeted CO_2 removal rate. The lean solvent CO_2 loading used in the PACT pilot plant for this case was 0.165. To find an optimum lean loading, a range of lean loading from 0.165 to 0.30 was studied. Table 8 presents the required solvent flow rate calculated by the model for each lean loading to achieve 90 % CO_2 removal rate using the flue gas condition presented in Table 7.

Table 8. Required solvent flow rate to achieve 90 % CO_2 removal rate with the base-case flue gas composition with IMTP25 random packing material

Lean loading (mol CO_2/mol MEA)	Solvent flow rate (kg/h)	Liquid to gas ratio (L/G) (kg/kg)
0.165	340.7	1.32
0.18	363.4	1.41
0.2	400.8	1.55
0.21	420.3	1.63
0.22	447.7	1.73
0.23	475.3	1.84
0.24	508.7	1.97
0.25	549.2	2.12
0.26	601.1	2.32
0.28	752.3	2.91
0.3	954.4	3.69

The reboiler duty at each lean loading was calculated using the model. Then the specific regeneration energy requirement and the total equivalent work for each lean loading were calculated. As shown in

Figure 2, the minimum total equivalent work occurs at a CO_2 loading of 0.23. The specific regeneration energy requirement at this loading is 5.13 MJ/kg CO_2 to achieve a 90 % CO_2 removal rate, compared to the base-case with 0.165 lean loading, where the specific regeneration energy requirement is 7.1 MJ/kg CO_2. The nearly 15 % reduction in the specific regeneration energy requirement is associated with a nearly 39 % higher circulating solvent flow rate. Studying the absorber design performance suggests the absorber column is capable of handling the excess solvent flow rate. The additional operational cost associated with the increased pumping power is very small compared to the gain associated with the reduction in the steam requirement.

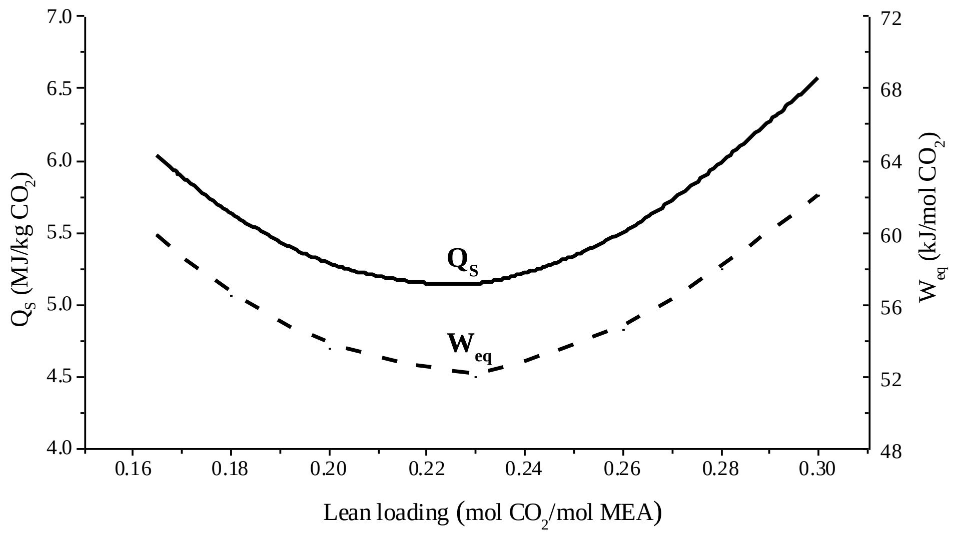

Figure 2. optimisation of lean loading for minimum total equivalent work with 125 kPa stripper pressure, 20 °C LMTD in cross heat exchanger, and IMTP25 random packing material

Although changing the lean loading to a higher value resulted in reducing the specific regeneration energy, the pilot plant energy performance is still sub-optimal and requires further modifications.

4.2.2. Cross heat exchanger

The rich solvent inlet temperature to the stripper is determined by the performance of the cross heat exchanger. This performance can be defined using the log mean temperature difference (LMTD) concept. In general, a lower LMTD is associated with higher capital cost for a given heat load, and the pilot plant cross heat exchanger currently operates with a 20 °C LMTD. To evaluate the extent to which a better performing heat exchanger will improve the plant energy performance, three different heat exchanger design specifications were analysed, corresponding to 20, 10 and 5° LMTD. Figure 3 shows the variation of specific regeneration energy requirement and total equivalent work with lean loading when the stripper column operates at 125 kPa.

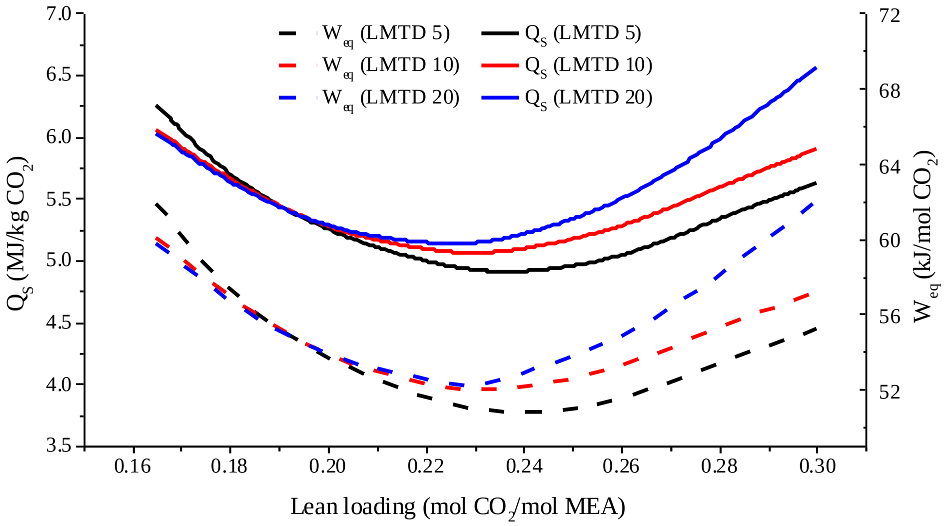

Figure 3. Specific regeneration energy requirement and total equivalent work variations with lean loading with 125 kPa stripper pressure, and IMTP25 random packing material, with 5, 10, 20 °C LMTD in cross heat exchanger.

The results show that the plant energy performance improves by up to 14 % across the range of lean loading by lowering the LMTD from 20 to 5 °C. Comparing the plant energy performance at the optimum lean loading, i.e. 0.23, suggests that having a 5 °C LMTD across the cross heat exchanger results in approximately 5 % reduction in the solvent regeneration energy requirement with almost 13 °C increase in the rich solvent temperature at the stripper inlet in relation to the base case with 20 °C LMTD. These findings suggest one way to improve the pilot plant energy performance is by replacing the cross heat exchanger with a high performing heat exchanger designed to operate with 5 °C LMTD. However, this benefit is associated with an additional cost of acquiring a larger heat exchanger. The studies discussed in the following sections are performed assuming the cross heat exchanger operates with a 5 °C LMTD.

4.2.3. Stripper operating pressure

It is possible to increase the stripper operating pressure and therefore its operating temperature by increasing the reboiler operating temperature via increasing the pressure of heat source, e.g. the boiler pressure (5). Currently the stripper operating pressure is 125±5 kPa when measured at the top of the column, and it was designed to operate at pressures up to 300 kPa. Figure 4 shows the effect of varying the stripper pressure from 125 to 250 kPa on total equivalent work and specific regeneration energy requirements across the range of lean loading from 0.165 to 0.30 assuming 90 % CO_2 removal rate, 5 °C LMTD at the cross heat exchanger, and 5 °C temperature approach across the reboiler.

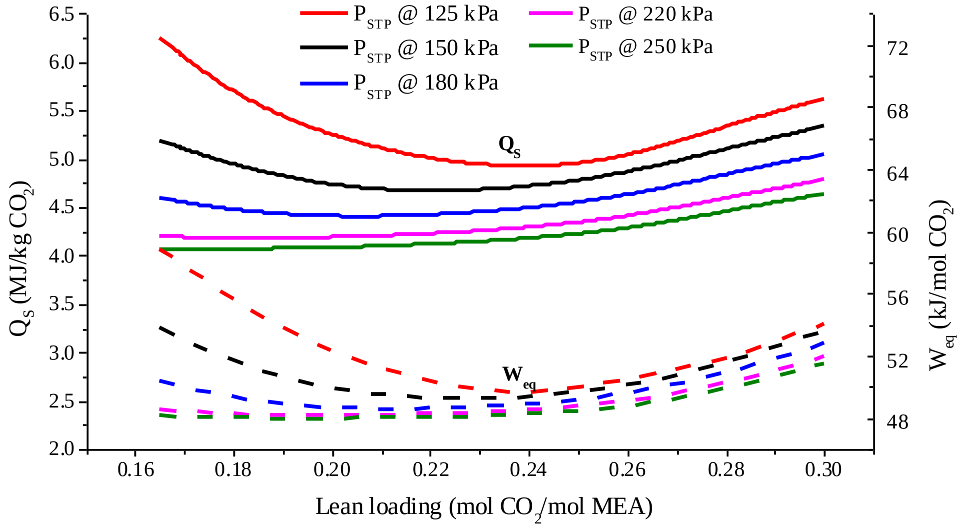

Figure 4. Specific regeneration energy requirement and total equivalent work variation with lean loading at various stripper operation pressure (125 kPa (red), 150 kPa (black), 180 kPa (blue), 220 kPa (magenta) and 250 kPa (green)) with 5 °C LMTD in cross heat exchanger, 5 °C temperature approach across the reboiler, and IMTP25 random packing material.

As shown in Figure 4, the specific energy requirement reduces with increasing the stripper pressure. Increasing the operating pressure from 125 to 250 kPa is associated with nearly a 17 % reduction in the specific regeneration energy consumption at their optimum lean loading. Operating at higher pressures in general reduces the CO_2 compression energy requirement although this is not considered for this pilot plant energy study. It appears increasing the stripper operating pressure is a meaningful way to enhance the pilot plant energy performance. However, increasing the pressure will increase the solvent temperature at the reboiler and throughout the column. The thermal degradation of MEA occurs mainly in the stripper packing and reboiler due to exposure to high temperature (36). Davis and Rochelle (36) studied the thermal degradation of MED and indicated that thermal degradation is minor when the solvent temperature at reboiler temperature is held below 110 °C but it accelerates above 130 °C. Figure 5 shows the variation of the solvent temperature at the reboiler with the stripper operating temperature. By considering a degradation threshold of 120 °C, based on data provided in Figure 5, 180 kPa pressure appears to be the most suitable operating pressure in order to gain benefits by operating the stripper at higher pressure and avoid a higher risk of solvent degradation and minimise corrosion problems.

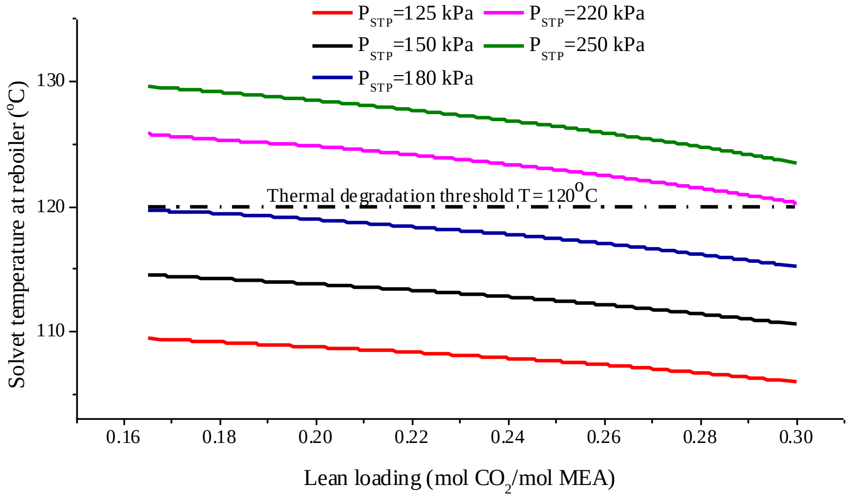

Figure 5. the variation of solvent temperature at the reboiler section with lean loading at various stripper operation pressures (125 kPa (red), 150 kPa (black), 180 kPa (blue), 220 kPa (magenta) and 250 kPa (green)) with 5 °C LMTD in cross heat exchanger, 5 °C temperature approach across the reboiler, and IMTP25 random packing material.

The lean loading at which the total equivalent work is minimised when the stripper operates at the pressure of 180 kPa is 0.21, provided a 5 °C LMTD in the cross heat exchanger and a 5 °C approach temperature across the reboiler. The solvent temperature at the optimum lean loading is 118.7 °C with the specific regeneration energy requirement of 4.4 MJ/kg CO_2. This amount of specific regeneration energy requirement is nearly 28 % lower than what has been currently recorded from the pilot plant operation. Table 9 summarises the proposed operating conditions to improve the energy performance of the PACT pilot plant to achieve 90 % CO_2 removal rate using IMTP25 random packing in all packed columns.

Table 9. Summary of proposed operating conditions for optimum operation of the PACT pilot plant to achieve 90 % CO_2 removal rate from typical natural gas fired flue gases when using the IMTP25 random packing

parameter	specification
Packing material	IMTP25 random packing
Flue gas temperature at absorber inlet	40 °C
Liquid to gas ratio	1.64 (kg/kg)
Lean solvent temperature at absorber inlet	40 °C
Lean loading	0.21 (mol CO_2/mol MEA)
Stripper pressure	180 kPa
Cross heat exchanger LMTD	5 °C

Reboiler approach temperature	5 °C

4.2.4. Packing material

It may not be fully advantageous to find conditions to optimally operate a CO_2 capture plant if is not associated with an efficient packing material. There are in general two different types of packing materials used in a CO_2 capture processes: random packing and structured packing. The pilot plant used for the case study is currently packed with the IMTP25 random packing because of ease of installation and its lower costs (28). Difficulties to achieve uniform distribution at the outset and the risk of maldistribution close to the column wall are problems typically reported for random packing, while structured packing materials are specifically designed to avoid such problems (37). Compared to random packing, structured packing has in general better mass transfer efficiency, good wettability and lower pressure drop (38). To further improve the energy performance of the PACT pilot plant with the fixed absorber design, i.e. height and diameter, and CO_2 removal rate, the current packing material should be replaced by a more efficient and better performing packing material from structured packing categories, such as Sulzer Mellapak 250Y. This modification will result in a reduction in the amount of circulating solvent required to achieve 90 % removal rate for a given lean loading due to the improved mass transfer efficiency in the absorber column. The lower solvent flow rate will therefore require less stripping steam to regenerate, as well as better performance of the stripper column itself by changing the packing material. All these will lead the pilot plant to operate with lower specific generation energy requirement. Table 10 summarises the solvent flow rate required to achieve 90 % CO_2 removal rate for the range of lean loading with the base-case flue gas compositions when replacing all the packing with the Sulzer Mellapak 250Y structured packing.

Table 10. Required solvent flow rate to achieve 90 % CO_2 removal rate with the base-case flue gas composition with Sulzer Mellapak 250Y structured packing, and the comparison with those for the IMTP25 random packing material

Lean loading (mol CO_2/mol MEA)	Lean solvent flow rate (kg/h)		Reduction in required solvent flow rate (%)
	Mellapak 250Y	IMTP25	
0.165	283.2	340.7	16.9
0.18	297.6	364.5	18.3
0.2	319.3	401.3	20.4

0.21	331.0	420.3	21.2
0.22	344.2	447.7	23.1
0.23	358.5	475.3	24.6
0.24	373.8	373.8	26.9
0.25	390.5	390.5	29.2
0.26	408.9	408.9	32.1
0.28	452.4	452.4	39.8
0.3	509.9	509.9	46.7

As presented in Table 10, the significant reduction in the required solvent flow at higher lean loading confirms the poor mass transfer efficiency of random packing at higher liquid to gas ratios. When using the Sulzer Mellapak 250Y structured packing, the simulation results also confirmed the stripper operating pressure of 180 kPa is the best option in terms of energy performance with respect to a 120°C thermal degradation threshold. Figure 6 shows the variation of total equivalent work and specific regeneration energy requirement with lean loading when using the Sulzer Mellapak 250Y structured packing with the stripper pressure of 180 kPa, 5 °C LMTD in the cross heat exchanger and 5 °C temperature approach at the reboiler. The curves related to the IMTP25 random packing with similar operating conditions were added for comparison.

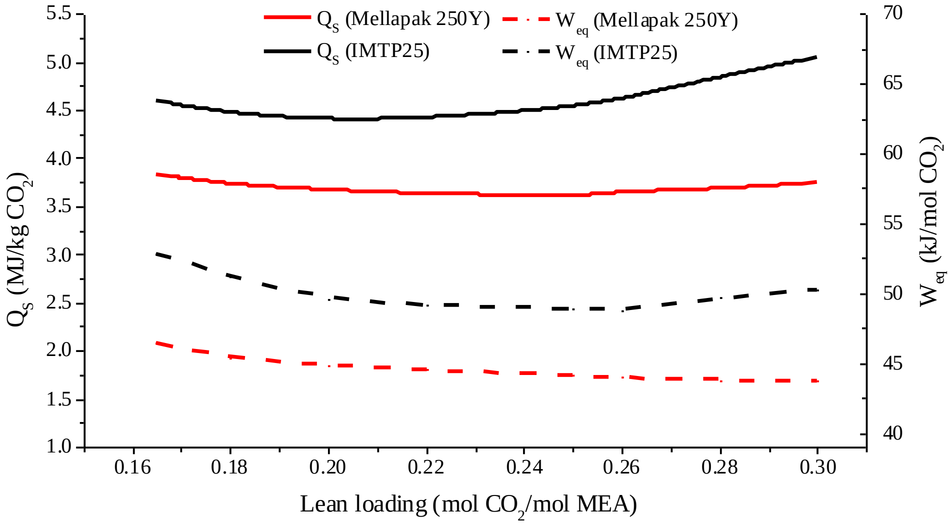

Figure 6. Optimisation of the lean loading for minimum total equivalent work and the specific regeneration energy requirement with the Sulzer Mellapak 250Y structured packing (black) and the IMTP25 random packing (red) to achieve 90 % CO_2 removal rate with the stripper pressure of 180 kPa

The minimum total equivalent work occurs at lean loading of 0.26 with a specific regeneration energy requirement of 3.64 MJ/kg CO_2, implying a nearly 39 % reduction in the specific regeneration energy requirement when compared with the current pilot plant operating condition to achieve 90 % CO_2 removal rate. The highest solvent temperature at the reboiler at the optimised lean loading is 107 °C. The specific regeneration energy requirement after changing the packing type is suitability within the industry range of 3.2 to 4.2 MJ/kg CO_2. The optimum operating condition using the Mellapak 250Y structured packing provides a 15 % reduction in the specific regeneration energy requirement compared to that provided by the optimum operating condition with the IMTP25 random packing. Table 11 summarises operating conditions to suitably improve the energy performance of the PACT pilot plant to achieve 90 % CO_2 removal rate for typical gas turbine flue gases when replacing all packing with the Sulzer Mellapak 250Y structured packing.

Table 11. Summary of the proposed operating condition for an optimum operation of the UKCCSRC/PACT CO_2 capture pilot plant to achieve 90 % CO_2 removal rate from typical natural gas fired flue gases when using the Sulzer Mellapak 250Y structured packing

parameter	specification
Packing material	Sulzer Mellapak 250Y structured packing
Flue gas temperature at absorber inlet	40 °C
Liquid to gas ratio	1.58 (kg/kg)
Lean solvent temperature at absorber inlet	40 °C
Lean loading	0.26 (mol CO_2/mol MEA)
Stripper pressure	180 kPa
Cross heat exchanger LMTD	5 °C
Reboiler approach temperature	5 °C

5. **Conclusions**

A rate-based model to simulate the CO_2 capture process using an aqueous solution of 30 wt. % MEA as solvent has been developed in Aspen Plus® Version 8.4 and validated using results of 5 experimental studies carried out at the UKCCSRC/PACT pilot plant in Sheffield, UK. The developed model was then used to assess the performance of the pilot plant in terms of energy consumption, and to propose new operating conditions to operate the pilot plant optimally in future. A number of performance parameters have been identified and varied for a given range of lean solvent CO_2 loading from 0.165 to 0.30 (mol CO_2/ mol MEA) to evaluate their effects on the plant energy performance. Two sets of operating conditions with two different packing materials were finally suggested to improve the pilot plant energy performance.

For the pilot plant to efficiently achieve 90 % CO_2 capture from flue gases with 5.5 % CO_2, typical of a natural gas fired applications, the following modifications were suggested:

- A more efficient cross heat exchanger has the potential to improve the stripper performance by providing the rich solvent with a temperature closer to its bubble point, also known as bubbling point, at the stripper inlet. Simulation results showed a nearly 5 % reduction in the specific regeneration energy requirement associated with the rich solvent being heated up by further 13 °C when using a 5 °C LMTD cross heat exchanger instead of the current one with a 20 °C LMTD.
- Considerable energy savings can be achieved by increasing the lean loading level, provided that the absorber column is capable of operating at higher liquid rates, which is achievable for the case of the PACT pilot plant. Simulation results have shown that by solely increasing the lean loading from 0.165 to 0.23, with no other change of the pilot plant operating condition, the specific regeneration energy requirement was reduced by nearly 15 %. The additional cost associated with the 28 % increase in the solvent flow rate is insignificant compared to the energy gain realised in the regeneration process.
- The stripper operating pressure also has a significant effect on the regeneration energy performance. Simulation results showed that by increasing the stripper pressure from 125 to 180 kPa the specific regeneration energy requirement will reduced by 28 %. The optimum lean loading to realise this gain is at 0.21 with a 118.7 °C solvent temperature at the reboiler section, which is reasonably below the thermal degradation threshold of MEA solvents.
- An efficient and modern packing material can contribute to significantly improve the overall performance of the PACT pilot plant by providing higher mass transfer efficiency, lower pressure drop and more efficient liquid and gas distributions. Simulation results suggest replacing the existing packing material with higher performing structured packing, e.g. Sulzer Mellapak 250Y will result in a nearly 40 % reduction in the specific regeneration energy when compared with the plant existing conditions. The proposed operating condition with the

Sulzer Mellapak 250Y structured packing outperformed the condition proposed with the IMTP25 random packing by nearly 15 %.

The main conclusions of this work should also hold for other plants of this type that employ 30 wt. % MEA solution as solvent.

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