Fuel, Volume 88, Issue 12, 7th European Conference on Coal Research and Its Applications, December 2009, Pages 2455-2462

Dynamic modelling of CO_2 absorption for post combustion capture in coal-fired power plants

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Abstract

Power generation from fossil fuel-fired power plants is the largest single source of CO_2 emissions. Post combustion capture via chemical absorption is viewed as the most mature CO_2 capture technique. This paper presents a study of the post-combustion CO_2 capture with monoethanolamine (MEA) based on dynamic modelling of the process. The aims of the project were to compare two different approaches (the equilibrium-based approach versus the rate-based approach) in modelling the absorber dynamically and to understand the dynamic behaviour of the absorber during part load operation and with disturbances from the stripper. A powerful modelling and simulation tool gPROMS was chosen to implement the proposed work. The study indicates that the rate-based model gives a better prediction of the chemical absorption process than the equilibrium-based model. The dynamic simulation of the absorber indicates normal absorber column operation could be maintained during part load operation by maintaining the ratio of the flow rates of the lean solvent and flue gas to the absorber. Disturbances in the CO_2 loading of the lean solvent to the absorber significantly affect absorber performance. Further work will extend the dynamic modelling to the stripper for whole plant analyses.

Keywords: CO_2 Capture, Chemical absorption, Dynamic modelling, Coal-fired power plant

1. Introduction

Power generation from fossil fuel-fired power plants is the largest single source of CO_2 emissions [1]. Coal-fired power plants release twice as much CO_2 per unit of electricity generated than natural gas power plants. However, these power plants play a vital role in meeting energy demands as coal is relatively cheap. In addition, coal-fired power plants can be operated flexibly as mid-merit plants in response to changes in supply and demand [2]. With growing concerns about the environmental impact of such plants effective CO_2 emission abatement strategies such as Carbon Capture and Storage (CCS) are required for their continued use. One approach to CCS is post combustion capture which involves the separation of CO_2 from the flue gas stream after combustion occurs (Figure 1). Chemical absorption is well suited for separating CO_2 from streams with low concentration of CO_2 (10-15% by volume) typical of pulverized fuel power plants [3,4].

1.1 Motivation

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Several studies have shown that the energy requirement for solvent regeneration would have adverse effects on power plant efficiency [5-8]. These effects have been studied using various steady state models and techno-economic assessments. However there are several gaps in the understanding of the impact of post combustion capture on the operability of the power plant. For instance, would such power plants be able to effectively operate at varying loads? What modifications would have to be made to the conventional start-up and shutdown procedures? What implications would heat integration between the power plant and CO_2 capture facility have on their operation [9]? These questions can be addressed by studying the dynamic behaviour of such plants. To achieve this, accurate dynamic models of the power plant and the CO_2 capture facility are required.

1.2. Post combustion capture via chemical absorption

Chemical absorption involves the reaction of CO_2 with a chemical solvent to form a weakly bonded intermediate compound which may be regenerated with the application of heat producing the original solvent and a CO_2 stream [3,4]. Monoethanolamine (MEA) being a primary amine reacts with CO_2 to produce carbamate via sets of liquid phase reactions [7,10]. A review of various reaction schemes describing this process is available in literature [11,12].

Figure 1 describes one of the popular technologies proposed for post combustion capture. The facility consists of two main units – the absorber and stripper columns which are both packed columns. Flue gas from the power plant is contacted counter-currently with lean MEA solution in the absorber. MEA chemically absorbs the CO_2 in the flue gas. This leaves a treated gas stream of much lower CO_2 content. The solvent solution (now Rich MEA) is regenerated in the stripper column using steam derived from the power generation process. CO_2 from the top of the column is compressed and transported away while the lean (regenerated) MEA solution is returned to the absorber column completing the cycle.

1.3 Novel contributions and outline of the paper

This study focuses on the dynamic model development of the absorber of the chemical absorption plant. It offers what is thought to be a unique comparison of the accuracy of two approaches to modelling the absorption process– the equilibrium-based and rate-based approaches. Also dynamic analyses of some disturbances to the absorber performance were carried out. Two disturbances were considered:

- Decreasing flue gas flow supplied to the absorber from the upstream power plant, and
- Increasing the CO_2 loading of the lean MEA solution from the stripper of the capture facility.

A review of various approaches to modelling the chemical absorption process is described in section 2. Two approaches were compared – the equilibrium and rate-based approaches. The model development process was described in section 3. These models were validated with results from a pilot plant study [13] in section 4. Two dynamic scenarios were simulated

– reducing power plant load and increasing CO_2 loading of the lean solvent. The results are presented in section 5. Conclusions were drawn in section 6 and recommendations for future work were given in section 7.

2. Developments in modelling chemical absorption of CO_2

Post combustion capture with MEA is a reactive absorption process. Two main phenomena are involved: mass transfer of CO_2 from the bulk vapour to the liquid solvent and the chemical reaction between CO_2 and the solvent.

A number of studies have employed steady state models of the chemical (or reactive) absorption process at different levels of complexity. Kenig et al describes the different levels of complexity of these models as illustrated in Figure 2 [14].

The equilibrium stage models assume theoretical stages in which liquid and vapour phases attain equilibrium. These models may assume the reactions are at equilibrium or may consider the reaction kinetics. The rate-based approach is more appropriate in modelling reactive absorption processes since phase equilibrium is hardly attained in practice. At its lowest level of complexity, the chemical reactions of the rate-based model are assumed to be at equilibrium. A more rigorous approach involves the inclusion of an enhancement factor to estimate actual absorption rates (with chemical reactions) from known physical absorption rates. The enhancement factor is calculated based on estimated reaction rates and is best suited for processes involving single irreversible reactions. This approach has been employed by a number of authors in developing steady state absorber and stripper models [15-17]. At the highest level available, reaction kinetics are modelled directly. Models consider mass transfer resistances, electrolyte thermodynamics, the reaction system as well as the column configurations and provide a direct estimation of concentration and temperature profiles by implementing reaction rates directly into the transport and balance equations in the film and the bulk of the fluid. This approach has been applied to a number of cases [10,14,18,19].

This study compares the equilibrium stage with the rate-based models both with reaction equilibrium (bottom and top left models in Figure 2).

The dynamic behaviour of the CO_2 absorption process for post combustion capture using MEA has not been extensively studied. Schneider et al considered the dynamic simulation of purification of coke plant gases [18]. Kvamsdal et al considered the dynamic simulation of only the absorber of the process using dynamic models of reduced complexity. The model employs an enhancement factor based on the assumption of pseudo first-order reaction regime. Kvamsdal et al adjusted both inlet flue gas flow rates and absorber heights to match pilot plant performance. In this study only the inlet flue gas flow rates to the absorber were adjusted. Kvamsdal et al also assumed a constant value for the heat of reaction and vaporization of water [9].

3. Model development

This section describes the model development of the absorber using the equilibrium- and rate-based approaches.

3.1. Equilibrium-based approach

The equilibrium-based approach was implemented in Aspen Plus[1] based on its Radfrac column model. This model was steady state and assumes theoretical stages in which liquid and vapour phases attain equilibrium and perfect mixing occurs. To describe non-equilibrium processes, the performance of each stage is adjusted using an efficiency correction factor [17,18]. For simulation purposes, the specifications presented in Table 1 were used. In section 4, all equilibrium based results were obtained from Aspen Plus.

The physical property method used is the Electrolyte Non-random-two-liquid (NRTL) model with electrolyte inserts for MEA. This insert includes new parameters and Henry's constant for CO_2 in MEA.

3.1.1. Aspen Properties MEA solution chemistry

MEA electrolyte solution chemistry is used to predict the equilibrium mass fractions in the liquid and vapour phases. The following are the set of equilibrium reactions describing this chemistry [20]:

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

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

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

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

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

3.2. Rate-based approach

In the rate-based approach, actual rates of multi-component mass and heat transfer as well as chemical reactions are considered directly [21]. The mass transfer is described using the two-film theory using the Maxwell Stefan formulation. Heat and mass transfer resistances are modelled in the liquid and vapour films.

The rate-based model was developed from the Gas-Liquid Contactor model in Process Systems Enterprise's Advanced Model Library using their process modelling tool, gPROMS. With gPROMS, accurate dynamic models of processes can be developed as it is equation-based and inherently dynamic.

3.2.1. Model assumptions

The following assumptions were used in developing this dynamic model:

- Plug flow regime
- Linear pressure drop along the column
- No accumulation in liquid and vapour films as well as bulk vapour

[1] Aspen Technology Inc.

- Phase equilibrium at interface between liquid and vapour films
- Negligible oxygen content in the flue gas
- Negligible solvent degradation
- Negligible heat loss to the surroundings
- Liquid phase reactions

3.2.2. Material and energy balances

Material and energy balances are carried out on the bulk liquid and vapour.

Bulk Liquid: $$\frac{dM_i}{dt} = \frac{-1}{L \cdot A}\frac{\partial F_i^L}{\partial y} + N_i \cdot Sp \cdot MW_i \cdot \omega \quad (6)$$

$$\frac{\partial F_i^L\big|_{y=1}}{\partial y} = 0 \quad (7)$$

Where,

$$M_i = x_i \times M, \quad i = 1,...,n \quad (8)$$

$$\sum_{i=1}^{n} x_i = 1 \quad (9)$$

y is the axial position relative to the top of the absorber packing ranging from 0 (or top) to 1 (or bottom of the packing).

$$\frac{dU}{dt} = \frac{-1}{L \cdot A}\frac{\partial F_H^L}{\partial y} + Sp \cdot \omega\left(H_{liq}^{cond} + H_{liq}^{conv} + H_{abs}\right) \quad (10)$$

$$\frac{\partial T\big|_{y=1}}{\partial y} = 0 \quad (11)$$

Where

$$H_{abs} = N_{CO_2} \times h_{abs} \quad (12)$$

The specific heat of absorption, h_{abs} (J/mol), is estimated as a function of temperature and CO_2 loading based on expressions in literature [22]. F_H^L is the liquid enthalpy flow rate (J/s).

Bulk Vapour: $$0 = \frac{-1}{L \cdot A}\frac{\partial F_i^V}{\partial y} - N_i \cdot Sp \cdot MW_i \cdot \omega \quad (13)$$

$$\frac{\partial F_i^V\big|_{y=1}}{\partial y} = 0 \quad (14)$$

$$0 = \frac{-1}{L \cdot A}\frac{\partial F_H^V}{\partial y} + Sp \cdot \omega\left(H_{vap}^{cond} + H_{vap}^{conv}\right) \quad (15)$$

3.2.3. Mass transfer

Mass transfer was modelled with resistances in the liquid and vapour films. The diffusivity (χ) of CO_2 in the liquid phase was based on expressions provided by Vaidya et al [23]. The diffusivity (χ) of CO_2 and other components in the vapour phase was estimated using the Fuller's equation [24]. Mass transfer coefficients in the liquid and vapour films were determined by correlations given by Onda et al [25]. Molar fluxes (N_i) of each component were estimated using the Maxwell-Stefan formulation (applied to both liquid and vapour phases):

$$\frac{1}{\delta}\frac{\partial x_i^M}{\partial z} = \frac{1}{c_t}\sum_{k=1}^{n}\left(\frac{x_i^M N_k - x_k^M N_i}{\chi_{i,k}}\frac{\mu^R}{\mu}\frac{T}{298.15}\right) \quad (16)$$

Where, c_t is the total molar concentration in the phase, δ is the film thickness and x_i^M is the molar fraction.

3.2.4. Physical properties

The physical property estimation models were set up in Aspen Properties. Through the CAPE-OPEN Thermo interface, gPROMS can take advantage of Aspen Properties' extensive physical property database. The Electrolyte-NRTL properties method was selected. For simplicity, only four main components were considered – MEA, water, carbon dioxide and nitrogen (oxygen content was incorporated into the nitrogen composition). Aspen properties also includes electrolyte inserts for the Electrolyte-NRTL property method where electrolyte solution chemistry is accounted for [20]. The same physical property model was used in the equilibrium- based model was used here as well.

MEA electrolyte solution chemistry is used to predict the equilibrium mass fractions in the liquid and vapour phases at the interface. The same set of equilibrium reactions described in equations (1-5) is used.

4. Model validation

The models developed were validated using data from the Separations Research Program at the University of Texas at Austin. The absorber column of the pilot plant is a packed column with a diameter of 0.427m and total height of 6.1m. This column consists of two 3.05m packed bed sections with a collector plate and redistributor between the beds [13]. Out of the 48 experimental cases carried out in the research program, two cases (Cases 32 and 47) were selected for steady state validation purposes. These two cases were selected because of their relatively high and low liquid to gas (L/G) ratios respectively.

Table 2 shows the process conditions for the lean MEA and flue gas streams to the absorber while Table 3 shows some absorber column and packing specifications.

Simulation results were validated using the temperature profile of the absorber column measured in the pilot plant [13]. In addition the measured CO_2 loading of the amine solvent taken at different positions was compared with values obtained from simulation.

4.1 Validation and comparison of equilibrium- and rate-based models

4.1.1. Case 47

This case involved a relatively low liquid to gas (L/G) ratio thus a lower CO_2 capture level.

Because of the reported inaccuracy in the flue gas flow measurement [9,13], its value was adjusted to match reported capture levels as shown in Table 4. Both the equilibrium and rate-based models predicted lower rich solvent loading than what was measured while the absorption levels are virtually the same as measured.

The temperature profile in the absorber was used to validate the two models as shown in Figure 4. The rate-based model gives a slightly better prediction of the temperature profile. The equilibrium-based model predicts generally lower temperatures than what was measured.

4.1.2. Case 32

This case involved a relatively high liquid to gas (L/G) ratio thus a high CO_2 capture level.

Both models showed poor prediction of the temperature profile in the absorber. However, with further reduction in the inlet flue gas rate to 0.11kg/s, better predictions were observed as shown in Figure 6. However, this change implies higher CO_2 capture levels than what was measured in the pilot plant (Table 5).

These discrepancies (as seen in Table 6) may be due to the assumption that the reactions between CO_2 and MEA are at equilibrium as calculated by the electrolyte solution chemistry. Kinetically controlled reactions may therefore provide better predictions of the trend. The rate-based model still gives a better prediction of the absorber temperature profile (Figure 6) compared to the equilibrium based model. The equilibrium-based model predicts higher temperatures than those measured in the pilot plant study.

5. Dynamic analysis

These analyses consider the effect of disturbances on the performance of the absorber. Two scenarios are considered:

- Reducing power plant load – as a mid-merit power plant, power generation would not be continuously at base-load level. In this scenario, a 50% reduction in power plant load occurs.
- Increasing lean MEA solution loading – with disturbances in the stripper column operation, such as reduced reboiler duty, the CO_2 loading of the lean MEA supplied to the absorber may increase. This scenario involves a 10% increase in lean loading.

5.1 Reducing power plant load

In this scenario, the upstream power plant load was reduced from base-load (100%) to 50% load. It was assumed that the flue gas flow rate decreases correspondingly and the changes in component composition are negligible. This scenario was applied to Case 32. Two cases were considered:

- Case-A: Change of flue gas flow rate without changing liquid (solvent) flow rate
- Case-B: Change of flue gas flow rate with corresponding decrease in liquid solvent rate to maintain CO_2 capture level

The process was simulated with the base-load conditions (Case 32) for three minutes after which the above changes were implemented in ten minutes. Finally conditions were maintained for eight minutes.

The two cases are illustrated in Figures 7

5.1.1 Case-A

From Figure 8, the 100s curve represents the profile before dropping load. The other curves show a trend of increasing absorption levels with time. Since the flue gas flow rate is ramped down with time while the solvent flow rate is constant, an increase in L/G ratio occurs.

Figure 9 shows the change in the CO_2 absorption level as the flue gas leaves the absorber with the L/G ratio. CO_2 absorption levels increase almost linearly with L/G ratio up to ratios of about 8.0. Afterwards, the rate of increase reduces.

There is also a significant change in the temperature profile in the absorber as seen in Figure 10. The location of the temperature bulge gradually shifts toward the bottom of the column. Temperature values generally reduce as less quantities of CO_2 are absorbed.

5.1.2 Case-B

By reducing the lean solvent feed rate correspondingly (by 50%), roughly the same capture level and temperature profile (Figures 11 and 12) could be maintained through the period of change. This suggests that the absorption process is more sensitive to the L/G (liquid solvent to flue gas) ratio than their actual flow rates. Since the amount of steam required for regeneration corresponds to the amount of lean MEA circulated, the energy requirement of the stripper could be correspondingly reduced.

5.2 Increasing lean MEA solution loading

This scenario was applied to Case 32 (process conditions in Table 2). Conditions were maintained for three minutes. Then the CO_2 loading of the lean solution was ramped up by 10% (from 0.279 to 0.3069) within three minutes. Finally, conditions were then maintained for 30 minutes to achieve steady state. This is illustrated in Figure 13.

With increased CO_2 loading of the lean MEA supplied to the absorber, the CO_2 absorption level (Figure 14) drops from 94.4% to 85.5%. CO_2 absorption levels could be maintained by either increasing the flow rate of lean MEA solvent to the absorber or decreasing the CO_2 loading of the solvent. The latter can be achieved by either increasing the stripper's reboiler duty or adding fresh MEA solution from solvent makeup tanks. Increasing the total solvent flow rate by 10% only results

in a capture level of 92.3%. Figure 15 shows that the temperature profile in the column is also affected by this change. The temperature bulge moves towards the bottom of the column.

6. Conclusions

This paper presents a study of the CO_2 capture with MEA based on the dynamic modelling of the process. Two models (equilibrium- and rate-based models) have been developed and compared. The rate-based approach yielded better predictions compared with the equilibrium-based approach.

Dynamic analyses of dropping the upstream power plant load and the effect of increasing CO_2 loading in the lean solvent were carried out. Simulation results reveal that the absorber operation is more sensitive to the L/G ratio than the actual flow rates of the solvent and the flue gas. Increased CO_2 loading in the lean solvent resulted in significant reduction in absorber performance.

7. Future work

It has been shown that the rate-based mass transfer model improves the predictions of the absorption process compared to the equilibrium based model. Another rate controlling factor is the reaction chemistry. The kinetics of the chemical absorption process would be improved by replacing the set of equilibrium reactions in the interface by kinetic and equilibrium reactions in the liquid film (top right model in Figure 2). This should give a better prediction of the absorption rates.

The stripper column model would subsequently be developed and the entire CO_2 capture facility model would be linked to a coal-fired power plant model for whole plant analyses.

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Table 1 Specifications for Equilibrium-based model

Description	Value
Number of equilibrium stages	7
Type of packing	IMTP
Packing material	Metal
Packing Dimension (m)	0.038
Packing height (m)	6.1
Condenser	None
Reboiler	None
Physical Property Method	Electrolyte NRTL

Table 2 Process conditions for Cases 32 and 47

	Case 47		Case 32	
Stream ID	FLUE GAS	LEAN MEA	FLUE GAS	LEAN MEA
Temperature (K)	332.38	313.32	319.71	313.86
Pressure (10^5 kPa)	1.033	1.703	1.035	1.703
Total flow (kg/s)	0.158	0.642	0.13	0.72
L/G ratio	4.6		6.5	
Mass-Fraction				
H_2O	0.0193	0.6334	0.0148	0.6334
CO_2	0.2415	0.0618	0.2520	0.0618
MEA	0	0.3048	0	0.3048
N_2	0.7392	0	0.7332	0

Table 3 Absorber column and packing data

Description	Value
Column inside diameter (m)	0.427
Height of packing (m)	6.1
Nominal packing size (m)	0.0381
Specific area (m^2)	145
Wetted area ratio	0.79

Table 4 Case 47 Process Conditions

	Pilot Plant Measurements	Equilibrium-based model	Rate-based model
Lean Solvent loading (mol/mol)	0.281	0.281	0.281
Rich Solvent loading (mol/mol)	0.539	0.500	0.487
CO_2 Absorption level (%)	69	68.8	69.2
Flue gas flow rate (kg/s)	0.158	0.172*	0.172*

*Adjusted flue gas flow rate

Table 5 Case 32 Process Conditions

	Pilot Plant Measurements	Equilibrium-based model	Rate-based model
Lean Solvent loading mol/mol	0.279	0.279	0.279
Rich Solvent loading mol/mol	0.428	0.469	0.464
CO_2 Absorption level (%)	95	97.8	94.4
Flue gas flow rate (kg/s)	0.13	0.12*	0.12*

*Adjusted flue gas flow rate

Table 6 Case 32 Process Conditions

	Pilot Plant Measurements	Equilibrium-based model	Rate-based model
Lean Solvent loading mol/mol	0.279	0.279	0.279
Rich Solvent loading mol/mol	0.428	0.456	0.456
CO_2 Absorption level (%)	95	99.6	99.5
Flue gas flow rate (kg/s)	0.13	0.11*	0.11*

*Adjusted flue gas flow rate

Nomenclature

A	Cross sectional area (m^2)
c_t	Total molar concentration (mol/m^3)
F_i	Component mass flow rate (kg/s)
F_H	Enthalpy flow rate (J/s)
H	Enthalpy (J)
h	Specific Enthalpy (J/mol)
L	Height of column (m)
L/G	Liquid to gas
M	Mass Holdup (kg/m^3)
MW	Molecular weight (kg/mol)
N	Molar flux ($mol/m^2.s$)
n	Number of components
Sp	Specific area (m^2/m^3)
U	Energy Holdup (J/m^3)
x	Mass fraction
x_i^M	Molar fraction
y	Axial position
z'	Film position

Greek Symbols

δ	Film thickness (m)
μ	Viscosity (Pa.s)
ω	Wetted area ratio
χ	Diffusivity (m^2/s)

Subscripts

i	Component number
H	enthalpy
abs	Absorption

Superscripts

L	Liquid
V	Vapour
Lf	Liquid film
Vf	Vapour film
Lb	Liquid bulk
Vb	Vapour bulk
$cond$	Conduction
$conv$	Convection
I	Interface

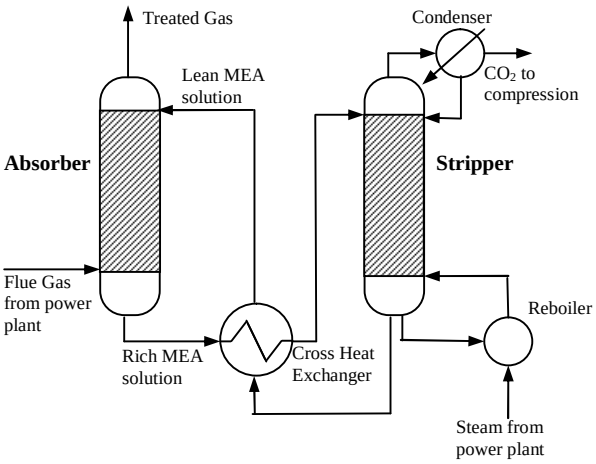

Figure 1 Simplified process flow diagram of Chemical Absorption process for post combustion capture from [9]

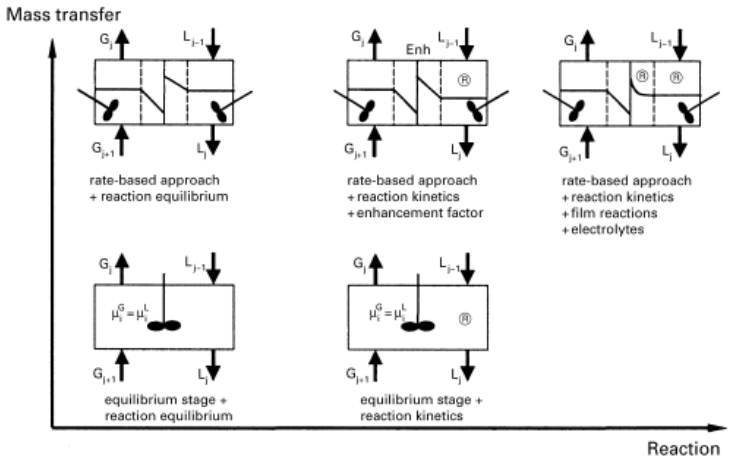

Figure 2 Different levels of reactive absorption model complexity from [14]

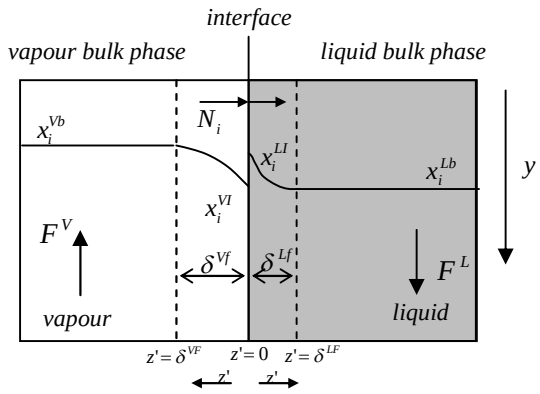

Figure 3 Liquid and vapour Bulks, films and interface adapted from [21]

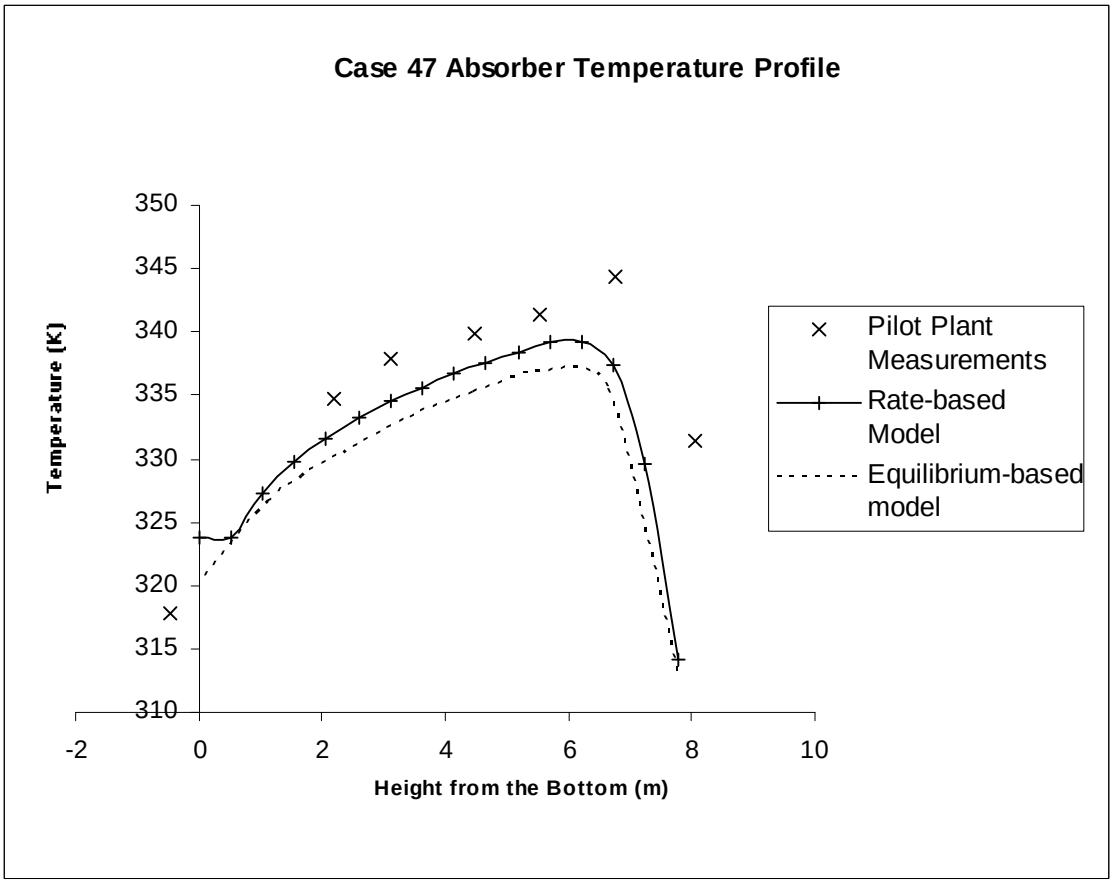

Figure 4 Absorber liquid temperature profile for case 47

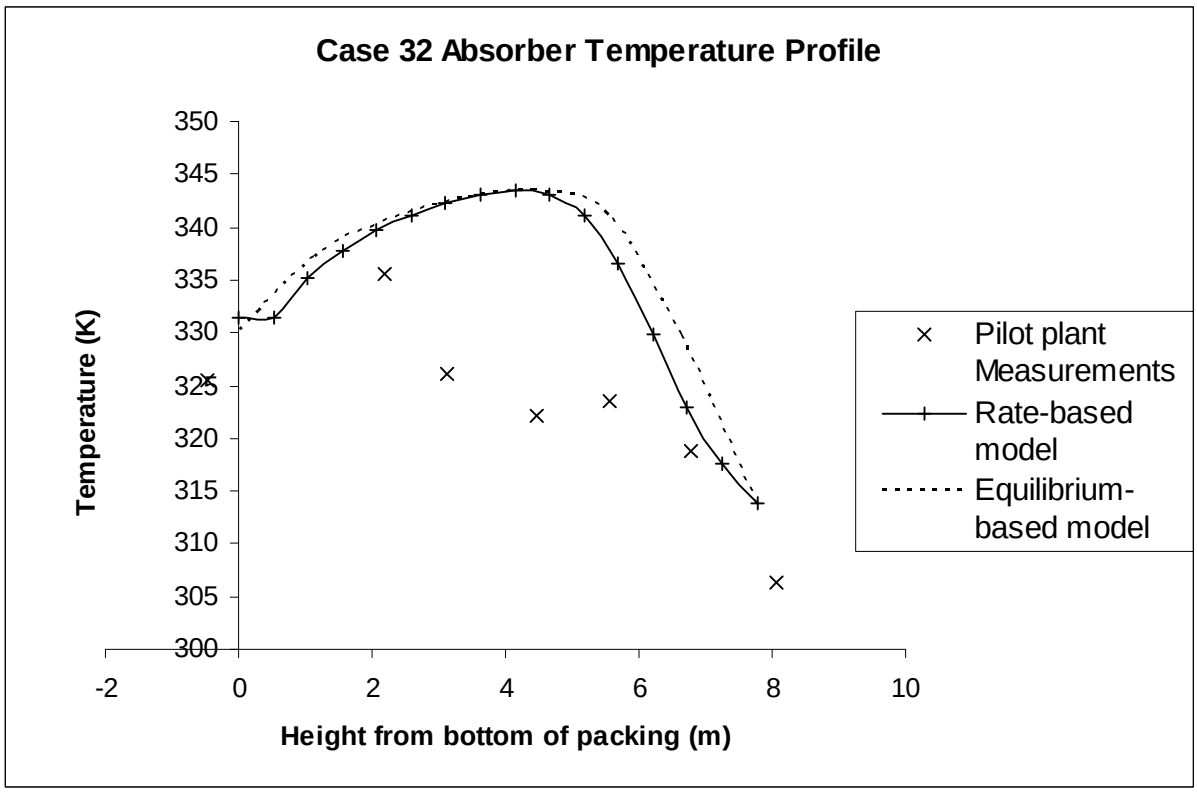

Figure 5 Absorber liquid temperature profile for case 32

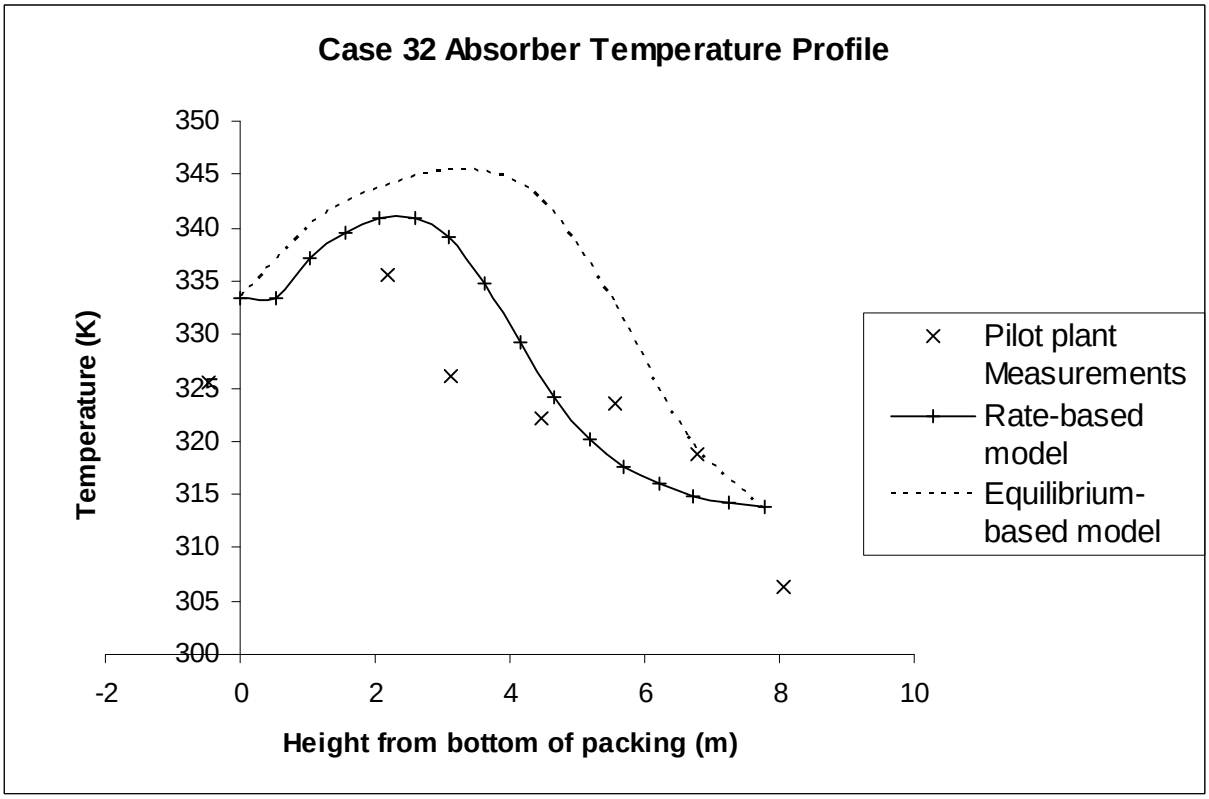

Figure 6 Absorber liquid temperature profile for case 32 with reduced flue gas flow

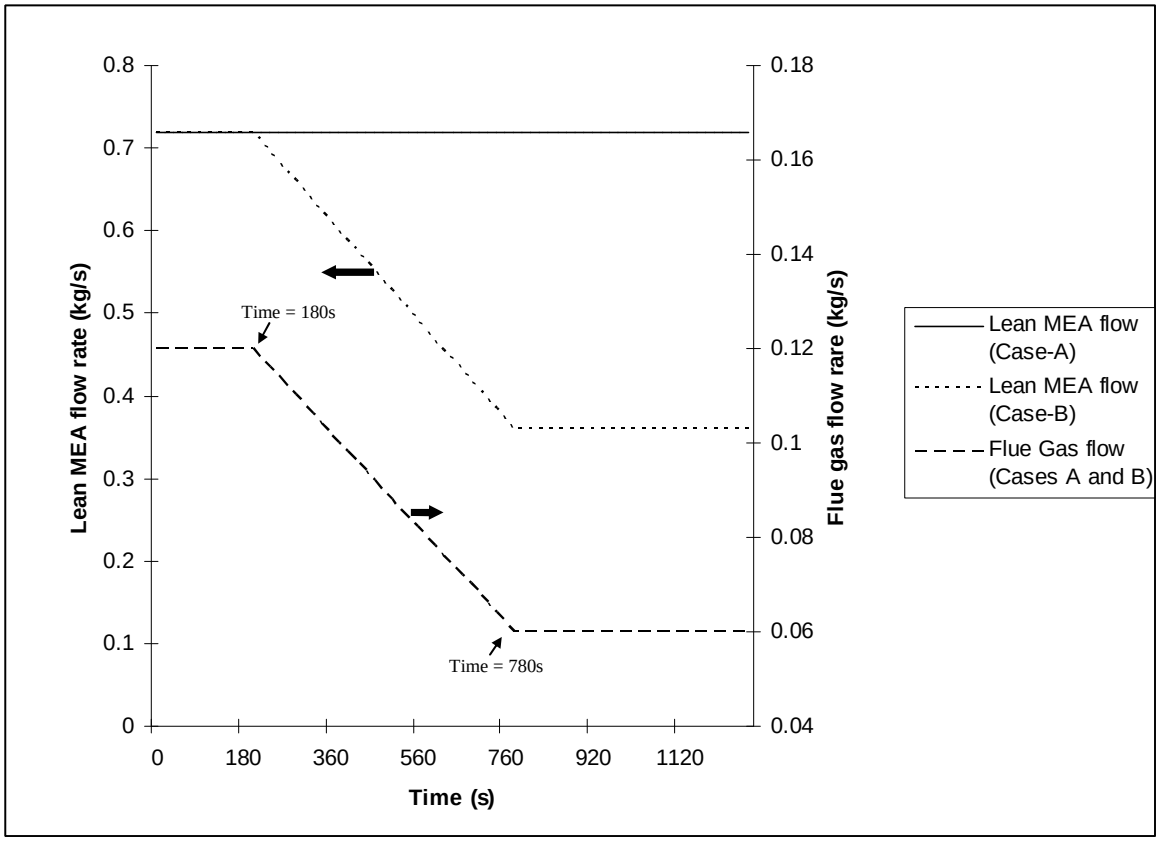

Figure 7 Flue gas and Lean MEA flow rate while reducing power plant load

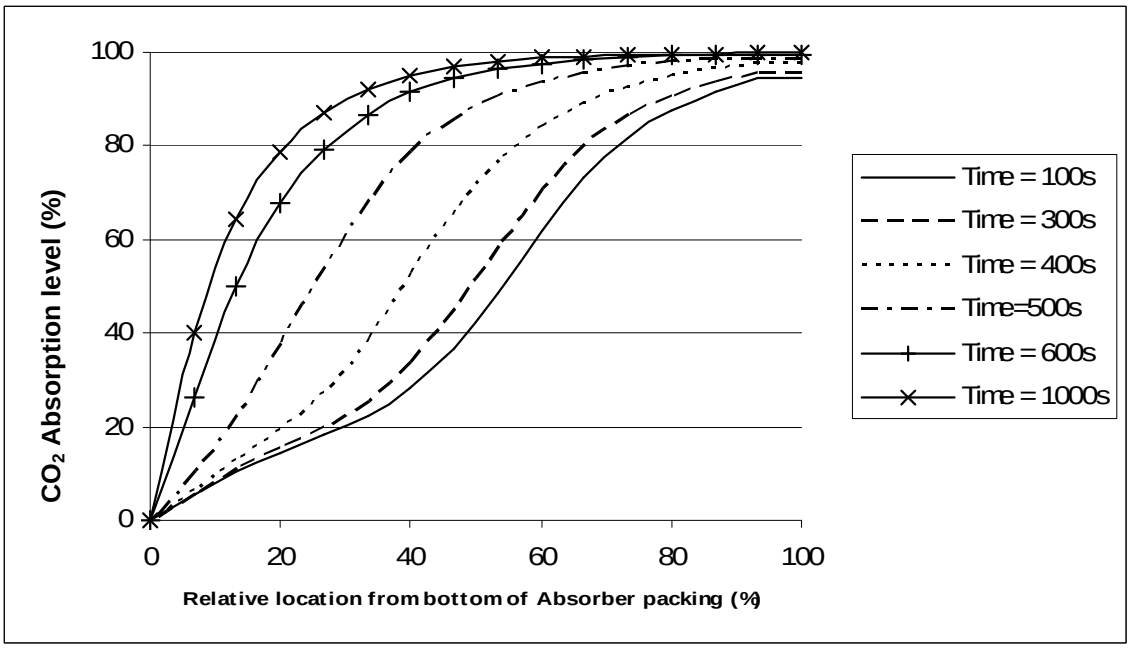

Figure 8 CO_2 absorption level – Case A

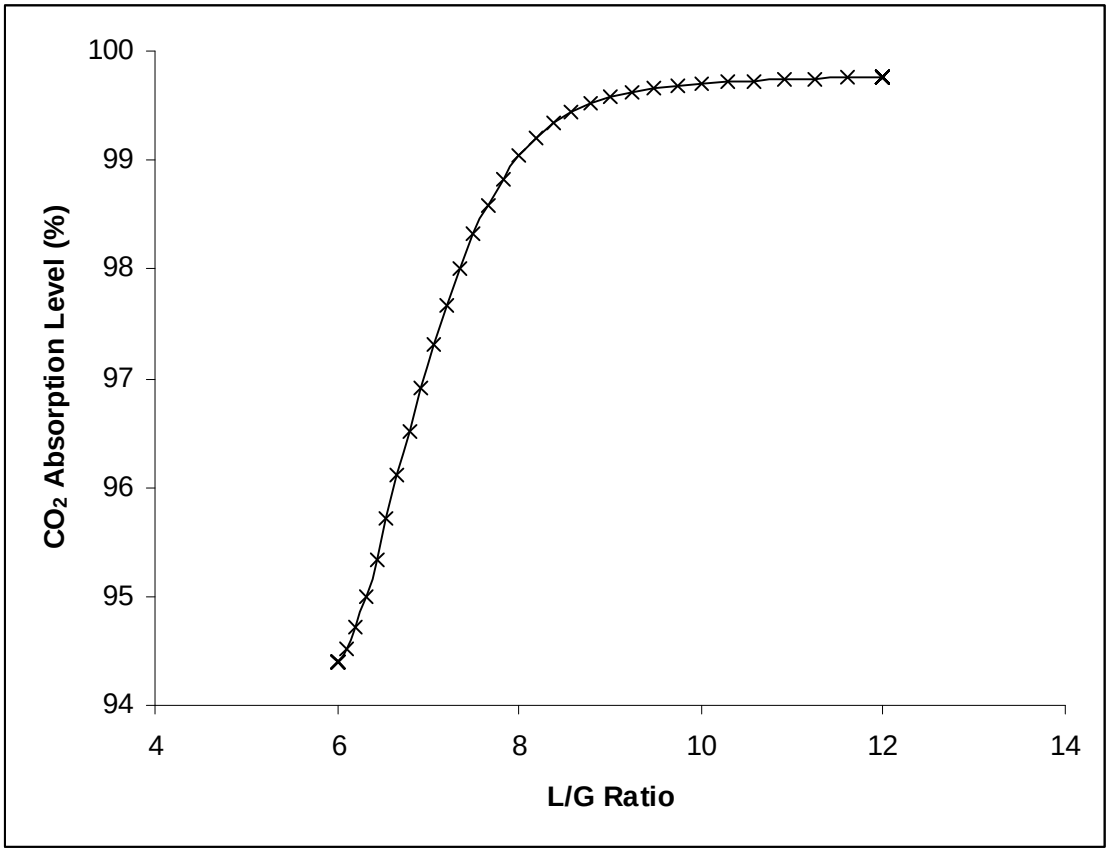

Figure 9 Change in CO_2 absorption levels with L/G ratio

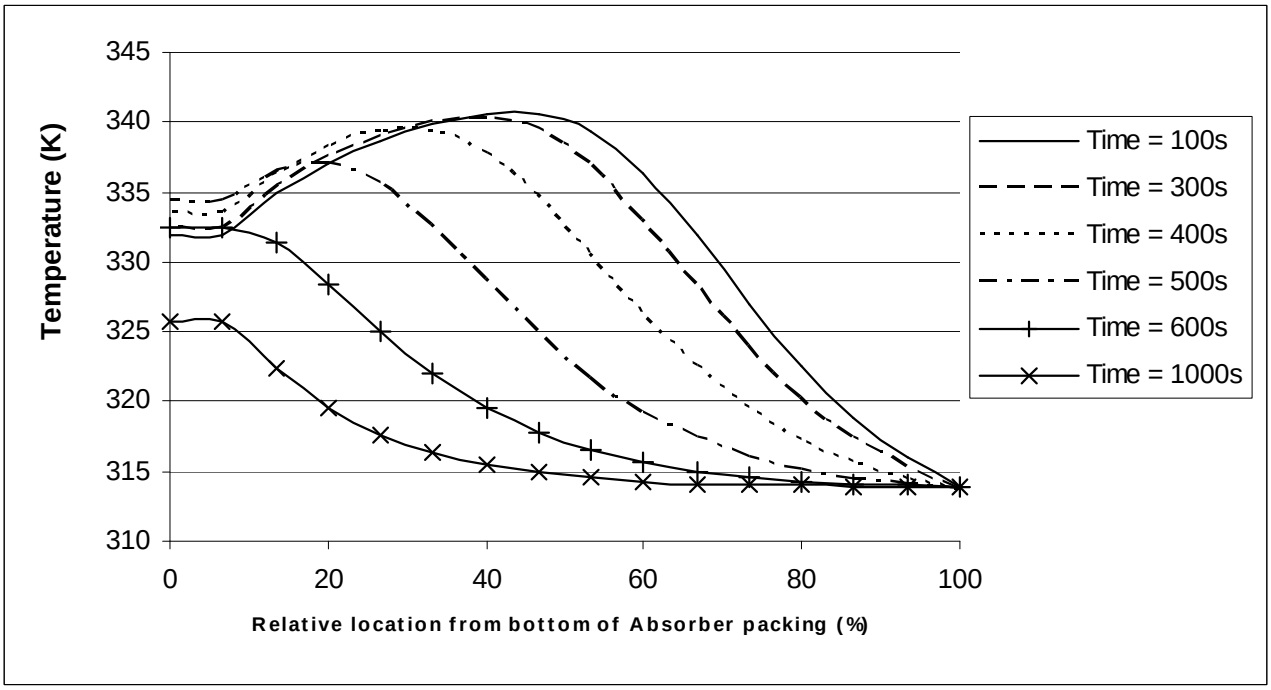

Figure 10 Temperature profile of absorber – Case A

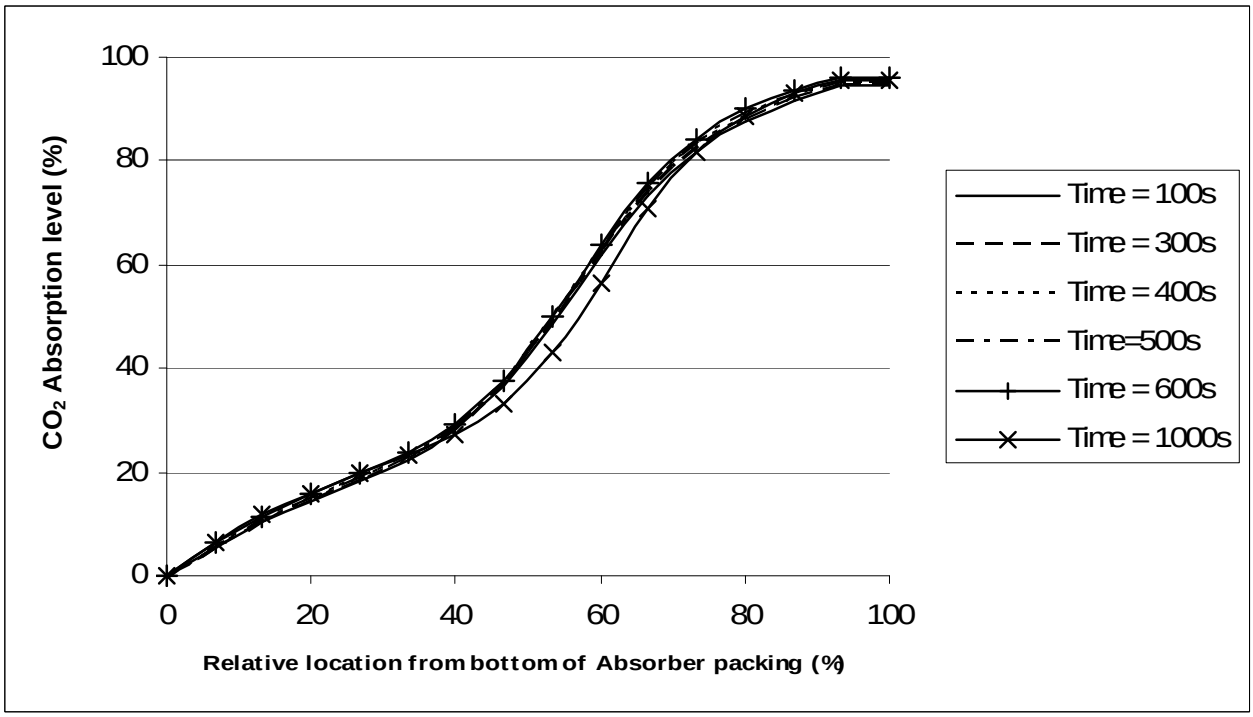

Figure 11 CO_2 absorption level – Case B

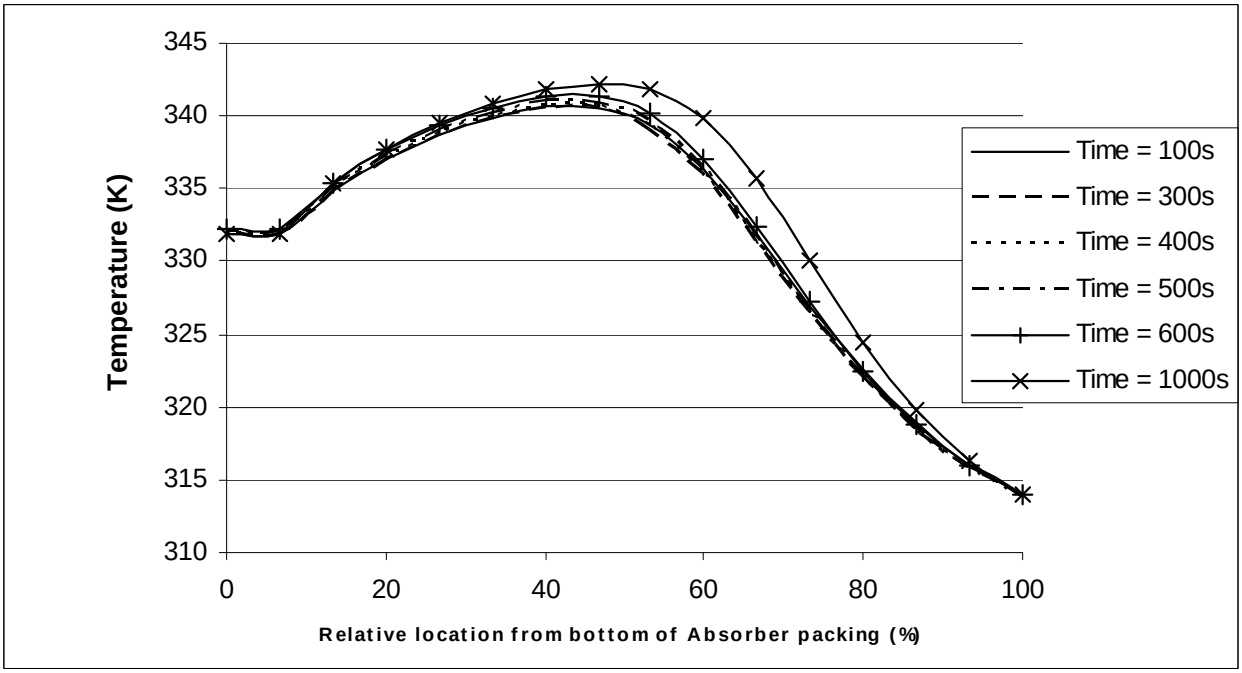

Figure 12 Temperature profile of absorber – Case B

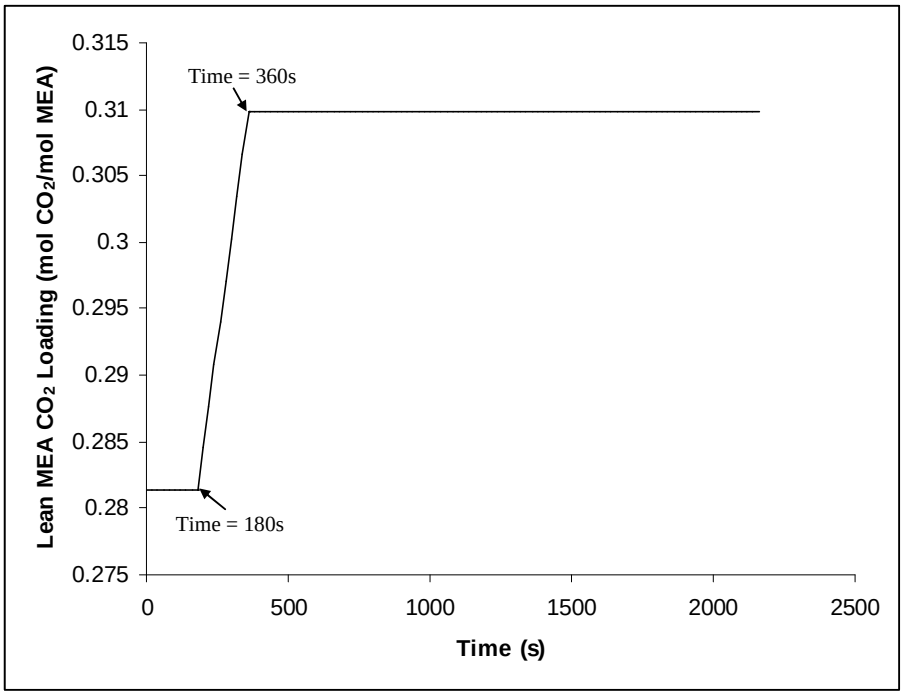

Figure 13 Increasing lean MEA CO_2 loading

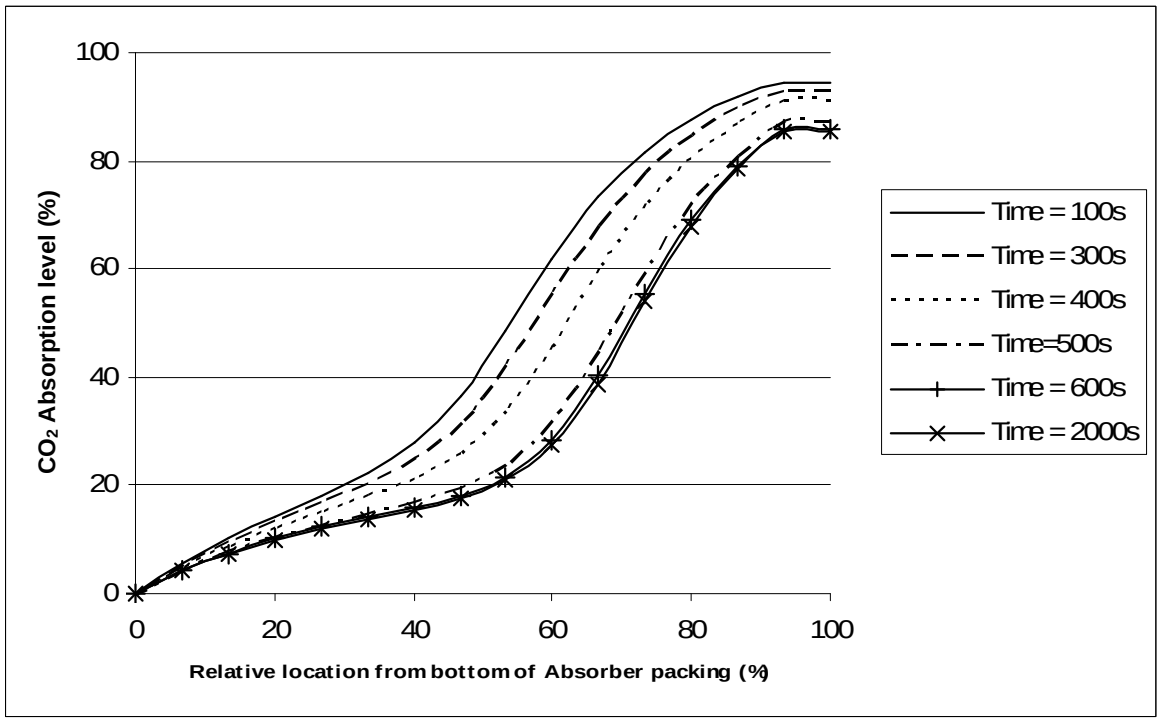

Figure 14 CO_2 absorption level while increasing CO_2 loading of lean MEA

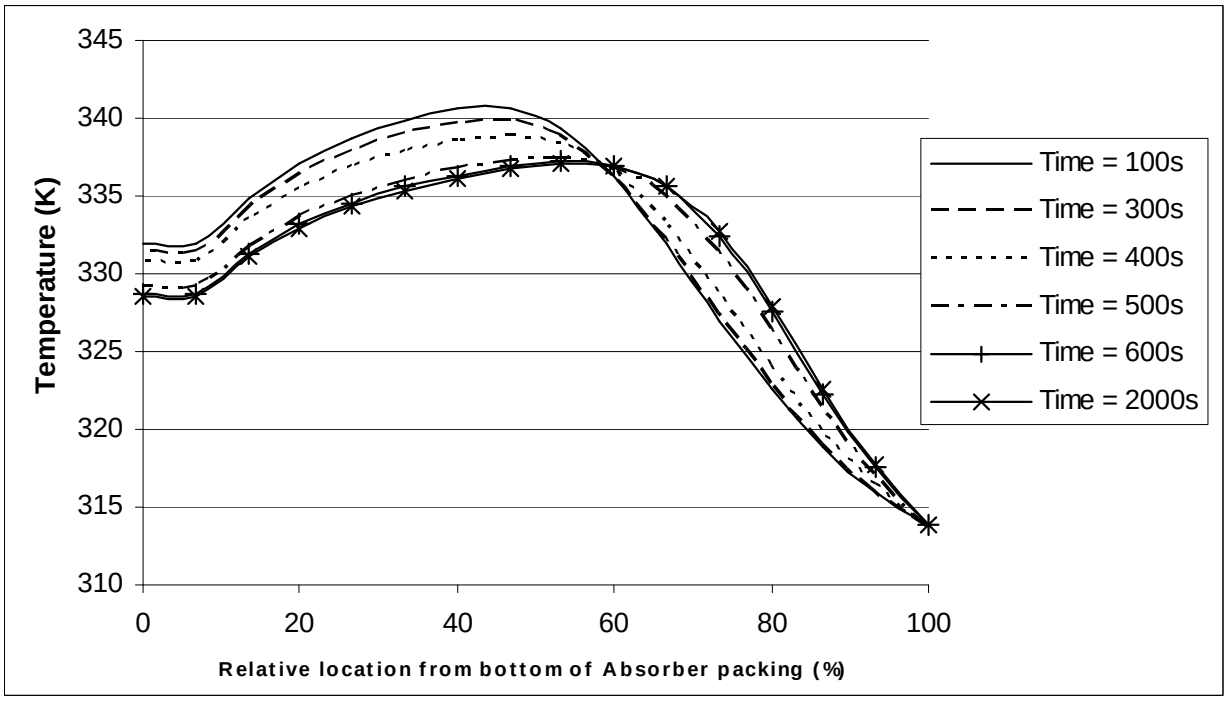

Figure 15 Temperature profile of absorber while increasing CO_2 loading of lean MEA