Prospects of using equilibrium-based column models in dynamic process simulation of post-combustion CO_2 capture for coal-fired power plant

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Abstract

This paper discusses the limitations and prospects of using equilibrium-based column models for the dynamic simulation of post-combustion CO_2 capture processes. Based on their features, one of three available commercial process simulators was chosen for this study. A pilot plant configuration adopted from literature was modeled and simulated using this simulator. Simulation results were compared with plant data and with results using standard rate-based models as available in literature. Temperature profiles in columns and overall mass and energy balances were found to be similar to plant data; however, CO_2 capture-rate, reboiler-duty, and rich-loading using the model were overestimated. A method of reduced stage efficiencies in the absorber was used, which improved performance prediction further with a maximum deviation of 5%. Further, this dynamic model was used to analyze the process subjected to variation in flue gas flow-rate with a similar trend of futuristic power plants by controlling either liquid to gas ratio or CO_2 capture-rate. Controlling liquid to gas ratio provided more control over the reboiler-duty while controlling the capture-rate focused on maintaining a certain capture ratio. The advantages and disadvantages of both methods are discussed and based on that, the controlling capture ratio was found suitable for using while power plant works flexibly with stringent emission regulations.

Keywords: Process modeling; CCS; Control structure; MEA; Transient analysis

1. Introduction

Fossil-fueled thermal power plants are among the largest point sources for anthropogenic CO_2 emissions. CO_2 capture and storage is considered as one of the options for reducing such emissions to attenuate the impact on the environment [1]. There are three main routes to capture CO_2 from power plants; post-combustion, pre-combustion and oxy-combustion processes, which are at different stages of development [1, 2]. Among them, the post-combustion CO_2 capture (PCC) processes using reactive absorption of CO_2 in flue gas from power plants are the most near-term technology [3]. The requirement for excess heat at reboilers in the stripper, the work requirement in the pumps to circulate the fluid through the PCC processes and the work requirement to compress the CO_2 will lead to an increase in the energy requirement of the power plant [4]. These, in turn, will reduce the overall power plant efficiency. As solutions to this, a number of techniques have been proposed in literature that aim to mitigate this reduction in efficiencies [5]. This requires extensive heat integration between the power plant and the PCC process and also within the PCC process itself, making the overall cycle configuration complex [6].

One of the major requirements for such integrated power plants is operational flexibility to cope with the continuous variation in electricity prices [7, 8]. In addition, with changing environmental policies, these complex plants need to be tuned to different CO_2 emission levels. In addition, optimization of shutdown and startup operations and risk and safety analysis need to be performed for stand-alone PCC plants before integrating them with new or existing power plants [7]. Understanding the performance of such plants during transient operation is the key requirement for identifying critical equipment in the plant, for designing the control structures and for predicting the modifications required under such conditions. Dynamic simulation is a useful tool for this purpose and numerous studies on dynamic modeling and simulations of such PCC plants have been reported in the literature as discussed in [6, 9, 10].

Rate-based column models built on two-film theory have been popularly used for absorbers and strippers, with a few exceptions where researchers have demonstrated use of equilibrium-based models in dynamic simulation [11-14]. Comparisons of the simulation results of rate-based and equilibrium-based models revealed the former to be more accurate and researchers recommended using such models [11, 15]. Rate-based models are more complex than equilibrium-based models, as they have a higher number of differential equations leading to longer computational time. In fact, the computational time has found to be approximately 35 times higher for rate-based models than for equilibrium-based models [13]. It is important to mention here that in most cases, highest order of accuracy in simulation results is not mandatory, and simpler models with known inaccuracies are acceptable [16].

Researchers have suggested simplifying the rate-based models to reduce the high time requirements [13, 16]; however, less is reported on how to improve the equilibrium-based models, other than the inclusion of Murphree stage efficiencies in the absorber model [11, 13, 14]. *Therefore, it is important to bridge the knowledge gap on equilibrium-based model limitations in dynamic process simulation and to identify ways to address the limitations.* This can help in addressing the issue of the computational time involved while using rate-based models and in finding a solution that is a suitable compromise between result accuracy and time required in dynamic simulations.

This work focuses on identifying the limitations in the use of equilibrium-based models for PCC processes and on finding possible solutions for those limitations. Once verified, the dynamic model can be used for analyzing PCC processes under various operational transients.

1.1 Objectives

Based on the above discussions the objectives of this paper are:

1. Identification of limitations in the use of equilibrium-based models in dynamic process simulation,

2. Investigation of methods to improve the performance prediction using equilibrium-based models,

3. Analysis of a dynamic model of a pilot plant under variable flue gas flow-rate conditions with two different control strategies maintaining the same liquid to gas ratio, and maintaining a constant CO_2 capture rate.

2. Methodology

As an alternative to the researchers programming their own code, a commercial process simulator with prebuilt unit operation models and avenues for the modifications and customization of such models is a better option for this study. Two of the main reasons for using a commercial process simulator are that they are widely used in the simulation of similar type of processes and that they have well verified and standardized models for equipment and thermodynamic properties.

2.1 Selection of process simulator

A number of simulators have been used for the dynamic simulation of PCC processes. They include simulators like Aspen Plus and Dynamics, Aspen Custom Modeller, gPROMS, MATLAB, UNISIM, and Dymola [9]. A few studies have simulated the power plant together with PCC process in dynamics using Aspen HYSYS [12], gPROMS, and Dymola [12, 17]. Validation of the simulation results using gPROMS has been performed using pilot plant data; however, no validation of simulation results using HYSYS is reported in the literature. Therefore, with the clear aim of selecting a commercial process simulator for use with an equilibrium-based model for both absorber and stripper, with a thermodynamic property data generation method and ease of customization, three available commercial process simulators were compared based on their features. The following table lists the features of all the simulators that have also been used previously for the simulation of such processes.

Table 1: Features of three available commercial process simulators

Parameter	Aspen HYSYS V8.6	Aspen Plus and Dynamics V8.6	gPROMS[1] [18]
Model type	Algebraic and ODEs with Space discretization	Algebraic and ODEs	Require modeling besides limited available standard models in library
Property data for amines	Yes (Acid gas)	Yes (MEA property package: E-NRTL)	Require external property data package/gSAFT
Property data for air/oxygen	Yes (Peng-Robinson)	Yes (Peng-Robinson)	Multiflash
Property data for fuel	Yes (Peng-Robinson)	Yes (Peng-Robinson)	Multiflash
CAPE-OPEN Thermo	Yes	Yes	Yes
Thermo-physical properties of materials	Yes (Customizable)	Yes	Require external property data package
Customization through	VBA	Fortran	Open software architecture via FOI, FPI, OCI, OSI
Interfacing with other software	In direct, in built interface with EXCEL	In direct, in built interface with EXCEL	In direct, in built interface with EXCEL, Open software architecture
Tools for parameter estimation	No	Yes	Yes

[1] gPROMS product family, not all the features are available with academic packages

Specific equipment models for power cycles	Standard model library	Standard model library	-
Advance control algorithms	Yes (DMC Plus, Sliding, MPC etc.)	Yes	Ease of interfacing with Matlab via EXCEL (FPI)/Control library
System identification tools	Yes (Artificial Neural Network)	Yes	Yes
Absorber/stripper model	Equilibrium based	Equilibrium as well as rate based using RateFrac in Steady state; In dynamics not possible to export the rate based model	Require modeling/Rate based in gCCS library
Novel component	Yes (property data in tabular form)	Yes	Models directly need to be attached
Solid handling	Yes (property data in tabular form)	Separate unit operation models	Yes
In-built performance curves for turbines	No	No	No
Numerical method	Only first order ADE solver	Higher order ADE solver	Finite difference and finite element methods
Dynamic optimization tools	No	Yes	Yes (MIO)
Ease of transfer from steady state to dynamics	From the same flow-sheet window	Exporting steady state case in dynamics	Need to identify parameters and specify before dynamic simulation
Spread-sheet	Yes	No	In built interface with EXCEL
Scheduler for implementation of control logic	Yes (Event scheduler)	No	In built interface with EXCEL

It may be observed from the table that almost all of the process simulators either have or can incorporate the required property data methods for both PCC and power plants. Equations of states (EOSs) for property data generation for amines are available with both the Aspen simulators. In gPROMS, property data or linearized curve fitted data need to be incorporated for the calculation of thermodynamic parameters, besides standard property libraries such as Multiflash and gSAFT [18]. In terms of process models, standard models with scope for customization are available in the form of model libraries in most of the simulators. This helps in eliminating re-engineering to formulate the basic models for all the equipment involved for this process. Further development of models and the required scope to incorporate them within the simulation is highly possible in these commercial process simulators; however, in terms of ease of customization, HYSYS and gPROMS were found to be the most acceptable. Continuous development of plant model libraries of different simulators is also reducing the effort involved to develop rigorous models. Therefore, any one of the three simulators can be used for our purpose and Aspen HYSYS V8.6 was selectted as the column models are based on equilibrium calculations and a widely accepted thermodynamic property-data generation method is available for simulating PCC processes.

2.2 Cycle configuration and equipment specifications

The first ever PCC process, which has been commercially deployed at the SaskPower coal power plant at Boundary Dam is yet to be delivered detail plant configurations and plant performance data that can be used for the study of model validations and verifications [19]. Therefore, it is difficult to obtain from literature full-scale plant data for the purpose of this study. A number of experimental pilot plants exists throughout the world. In this work, data from the pilot plant from the Separations Research Program at the University of Texas, Austin, USA as available in the literature have been used [20]. These data have also been used for the validation of dynamic models in [15, 21]. In their work, they used two sets of experimental data out of 48 trial runs in the pilot plant by [20] because of their relatively high and low liquid to gas (*L/G*) ratio. Similar sets of experimental data to those of trial run 32 and 47 (referred to as Case 32 and Case 47 hereafter in the paper) were used here. The process flow-sheet in this work was built based on the thermodynamic conditions mentioned in [15, 21]. The process conditions, equipment and the control specifications, and solution methodology of the models are presented hereafter in this subsection.

The pilot plant configuration was adapted from [20] as shown in Figure 1. This plant uses primary amine, MEA as solvent, whereas, the flue gas stream consists of nitrogen, water and CO_2. Two cylindrical columns with 6.1 m of packing with random packing materials were used as absorber and stripper. Individual state points of the process were either adopted from [15, 21] or estimated using the process simulator and they differ for the two cases selected for this study. The estimated individual state points are mostly near the stripper, as this is not defined in detail in the above mentioned literature. Thermodynamic conditions for some of the state points used in this work are presented in Table 2.

The following assumptions were made while estimating the process conditions in steady state:

1. No pressure drops in heat exchanger, cooler were considered.
2. The stripper works with constant pressure.
3. Piping and corresponding pressure drops were ignored.
4. Heat transfer to and from the atmosphere was neglected.
5. The minimum approach in the heat exchanger was considered as 15^0C.
6. The adiabatic efficiencies of both lean and rich MEA pumps were considered as 75%.
7. The stripper equilibrium stage efficiency was considered as 1.00.

In the dynamic simulation of the plant, the following were assumed:

1. Piping was ignored and corresponding pressure drops were included in neighboring equipment.
2. An additional lean MEA buffer was used to maintain lean loading after the rich-lean heat exchanger. The additional pump was used to maintain the inlet liquid pressure of the absorber. The loss of MEA

mostly occurred at the absorber and it was found to be vented with the treated gas, as no water wash section was considered.

3. The condenser and reboiler were modeled as volumes with suitable heat duty.
4. The heat exchanger was modeled as plate-fin one and configurations and thermal properties were estimated based on the steady state conditions.
5. Pumps were specified by their efficiencies and duty. Mass flow rate was maintained by suitably controlling the duty considering it as supplied by the energy stream.
6. No sump was used along with the absorber in the process flowsheet.
7. Pressure gradient in the absorber and regenerator were considered to be same.

The equipment specifications are presented in Table 3 as adopted from [15, 21]. The equilibrium stages for both the absorber and the regenerator were estimated using equilibrium stage models in steady state, constraining them with CO_2 capture rate and optimum reboiler duty for absorber and regenerator, respectively. However, the dimensions of the condenser and the reboiler in the stripper were estimated with the volume provided.

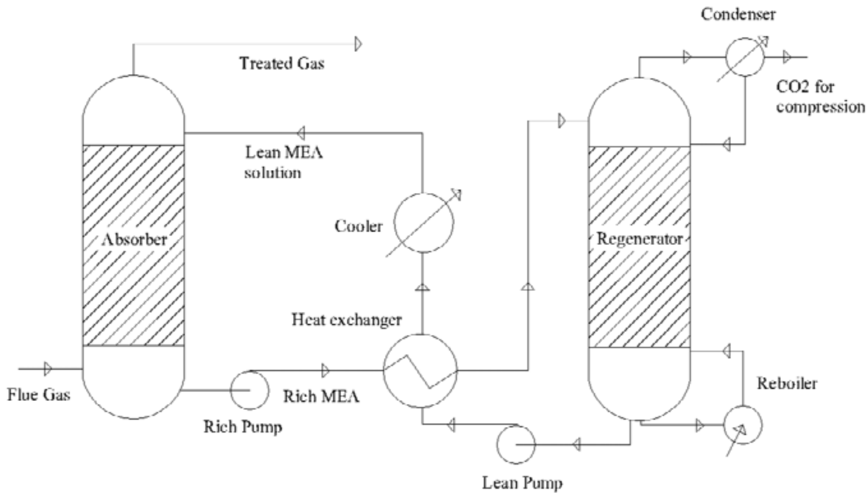

Figure 1: Typical process flow diagram of PCC process; the pilot plant configuration was adapted from [20] to fit

Table 2: Process condition for inlet streams to absorber for Case 47

Parameters	Flue gas		Lean MEA Solution		Rich MEA	
	Case 47	Case 32	Case 47	Case 32	Case 47	Case 32
Temperature (K)	332.38	319.71	313.71	313.86	356	358
Pressure (bar)	1.033	1.033	1.703	1.703	0.68	1.6
Mass flow rate (kg/s)	0.158	0.13	0.642	0.72	0.746	0.745
Mass fractions						
Water	0.0193	0.0148	0.6334	0.6334	0.6085	0.6122
CO_2	0.2415	0.2520	0.0618	0.0600	0.0966	0.0971
MEA	0	0	0.3048	0.3066	0.2943	0.2901
Nitrogen	0.7392	0.7332	0	0	0.0006	0.0006

Table 3: Specification of absorber and stripper

Parameter	Absorber	Stripper
Equilibrium stages	7	7
Type of packing	IMTP	IMTP
Packing material	Metal	Metal
Packing dimension	0.038 m	0.038 m
Packing height	6.1 m	6.1 m
Condenser volume	-	2 m^3
Reboiler volume	-	1 m^3
Column inside diameter	0.427 m	0.427 m
Specific area	145 m^2/m^3	420 m^2/m^3

2.3 Control structure

In order to maintain a constant rate of CO_2 capture in the plant, it was necessary for the condenser temperature, reboiler temperature, level and makeup water flow rate to be controlled [21]. However, controller for CO_2 capture rate was not considered; instead constant flue gas and lean amine mass flow-rate were specified. In addition, for the stability of the operation, the condenser level was also controlled. Therefore, five linear controllers were used in the simulation. PI-type controller, with relatively high proportional gain, were used for all the controllers. The table below presents the controller specifications. The relay-based auto-tuning method available with the process simulator was used to tune all of them.

Table 4: Controller specifications

Equipment	Control variable	Measured variable	Manipulated variable	Set point
Condenser	Condenser temperature	Vessel temperature	Heat duty	320 K
	Condenser liquid percentage level	Liquid percentage level	Bottom pump speed	40%
Reboiler	Reboiler temperature	Vessel temperature	Heat duty	388 K
	Reboiler liquid percentage level	Liquid percentage level	Rich pump speed	50%
Make up vessel	Water flow fraction at lean MEA	Water flow fraction at lean MEA	Pump duty	0.6334

2.4 Thermo-physical property data generation method

Use of the Amines Property Package was found to be suitable for MEA as it had also been used in [12, 22]. However, in the latest version of the simulator (V8.6), this property data package is merged with the Acid gas property generation method. Therefore, for our simulation, the Acid Gas Property Package was used and validated with the steady state simulations of the absorber as presented in a latter section.

Constant heat capacity and thermal conductivity data for metal and insulations were used as supplied. As no heat in-leak to the system or dissipation to the atmosphere was considered, insulation property data had no impact on the temperature profiles of the vessels (condenser, reboiler and make-up vessel) and heat exchangers.

2.5 Solution methodology

The Euler implicit method, a first order numerical method, was used for solving the models comprising sets of ordinary differential equations. The time required to solve a dynamic model changes inversely with the time step-size of the integrator. Therefore, a relatively higher time step-size of 2 s was used in all the dynamic simulations. The results of the simulation for the selected parameters were recorded after every 20 s. Additionally, as the flow-sheet needed to be initialized properly to avoid any divergence at the start of the simulation, initialization was performed by specifying each inlet, outlet streams and vessel wall of any equipment at atmospheric conditions as in the real plant. Pressure specifications in the boundary streams of the flow-sheet and flow specifications at the intermediate streams in the stripper were used.

3. Identification of limitations of equilibrium-based models

In order to identify the limitations of the equipment models, the property data generation method first needs to be verified first. Steady state simulation is one way of doing that, as the process simulator uses rate-based models of all the columns in steady state. The stand-alone absorber was simulated in steady state for verification of the accuracy of the property data generation method. Up on verification, the dynamic simulation of three cases – stand-alone absorber, stripper and complete plant model – was performed, where the column models were based on the equilibrium approach. It is important to validate the dynamic models against both the steady state and the transient plant operational data. Steady state operating data are available in the literature; however, there is a dearth of transient operational data in open literature. Therefore, in this paper, transient operational data generated using already validated rate-based models by Lawal et al. [23, 24] have been used. Therefore, pilot plant data [15, 20, 21] and simulation results from the rate-based model by Lawal et al. [15, 21] were used for comparison with the results obtained from the simulations of all three cases. As previously mentioned, two sets of experimental data from [20], Case 32 and Case 47, were used here. Temperature profiles of columns and the variation of different parameters under nominal operation of the plant were compared with the data mentioned above to identify deviations in the simulation results.

3.1 Verification of property data generation method using steady state simulation

The stand-alone absorber model consists of absorber and rich pump. Lean MEA and flue gas streams are the inlets to that model and treated gas and rich MEA streams are the outlets (see Figure 1). The steady state model of the absorber in the simulator is rate-based; therefore, this can help to distinguish the effects of the Acid gas property package on the simulation of the PCC processes. Two steady state flow-sheets based on two experimental cases, i.e. Case 32 and 47, were built with the specifications mentioned in Tables 2 and 3. These two cases were selected for their liquid to gas ratio. Case 32 is the highest among all the experimental runs with a *L/G* ratio of 6.5, whereas Case 47 is the lowest, with a *L/G* ratio of 4.6; thereby, this covers the range of *L/G* ratios for which the experimental results are available. It is also important to mention here that, as a result of selecting two extreme cases, the range of deviations can be identified and the maximum values obtained. These steady state simulation results of these two cases are shown in Table 5 and 6. This results were used for comparison. It is noteworthy that, due to inaccuracy in the flue gas flow rate measurement in the pilot plant, the flow rates were adjusted to match the reported capture levels of 0.11 kg/s instead of 0.13 kg/s for Case 32 and 0.172 kg/s instead of 0.158 kg/s for Case 47 [15].

Table 5: Comparison of steady state simulation results with experimental and rate-based simulation results for Case 32 (* user specified)

Parameter	Pilot plant	Rate-based simulation [15]	Current simulation	%-deviation from pilot plant data	Relative %-deviation from rate based model
Lean solvent loading (mol/mol)	0.279	0.279*	0.279*	-	-
Rich solvent loading (mol/mol)	0.428	0.456	0.429	-0.23	5.9
CO_2 absorption (%)	95	99.5	90.0	5.22	9.5
Flue gas flow rate (kg/s)	0.13	0.11*	0.11*	-	-

Table 6: Comparison of steady state simulation results with experimental and rate-based simulation results for Case 47 (* user specified)

Parameter	Pilot plant	Rate-based simulation [15]	Current simulation	%-deviation from pilot plant data	Relative %-deviation from rate based model
Lean solvent loading (mol/mol)	0.281	0.281*	0.281*	-	-
Rich solvent loading (mol/mol)	0.539	0.487	0.472	12.4	3.1
CO_2 absorption (%)	69	69.2	70.4	-2.0	-1.75
Flue gas flow rate (kg/s)	0.158	0.172*	0.172*	-	-

It may be observed from Tables 5 and 6, that there is negligible deviation for Case 32 and almost 12% deviation for Case 47 from the experimental data for rich solvent loading and approximately 5% deviation for Case 32 and a negative deviation of 2% for Case 47 in CO_2 absorption results. However, these results are found to be similar to those of the rate-based model, as is also evident from the tables, although, with an exception in capture ratio for Case 32. This confirms the use of the Acid Gas property generation method with respect to the Electrolyte-NRTL method obtained from Aspen Properties, which was used while simulating the rate-based model in [15].

Dynamic simulation cases for the absorber, stripper and the complete plant were developed using the Acid Gas package in the simulator. Temperature profiles for all the simulation cases were generated and compared as discussed in the following sections.

3.2 Simulation of stand-alone absorber operating in steady state

The flow-sheet containing only the absorber was subjected to dynamic simulation after specifying all the equipment sizes as per Table 3. Three different scenarios were simulated: Case 47, Case 32 and Case 32 with reduced flue gas flow rate. Temperature profiles for the absorber were generated (Figures 2, 3 and 4). It is to be noted that the pilot plant has a sump below the packing. The temperature of the sump wall was also measured there. However, as no sump was considered in the simulation, the reference point, “0”, was the bottom of the packing.

It may be observed that the temperature profile generated by dynamic simulation is better matched with the pilot plant data for Case 47, where the *L/G* ratio is less, when compared to Case 32. However, stand-alone models overall were not found to be as accurate as steady state simulation results and do not match exactly with the pilot plant data. Even compared to the rate-based model results, the predictions show more deviation for the simulation of Case 32 and Case 32 with reduced flue gas flow rate. The deviations in temperature predictions at the bottom most position of the absorber led to a different temperature at the outlet as may be observed in all the figures. This changed the stream temperature of rich MEA and consequently may affect the inlet temperature to the stripper and thereby its performance. Higher temperature at the top of the absorber predicted higher mass transfer vis-à-vis higher absorption rate, thereby resulting in deviation of the CO_2 capture level as observed in the simulation. Our simulation predicted a 99.9% rate of CO_2 absorption, whereas the rate-based model predicted close to 97%, while it was found to be 95% in the pilot plant. This difference, however, did not greatly affect the overall mass balance in the cycle and the trends match those of the pilot plant.

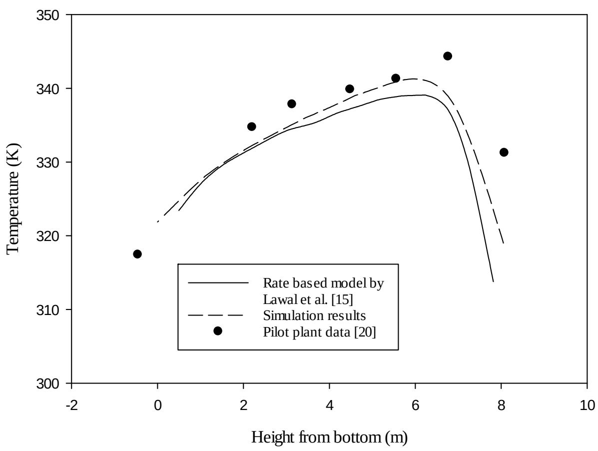

Figure 2: Comparison of absorber temperature profile in dynamic simulation with pilot plant [20] for Case-47 and with simulation results of Rate-based model by [15]

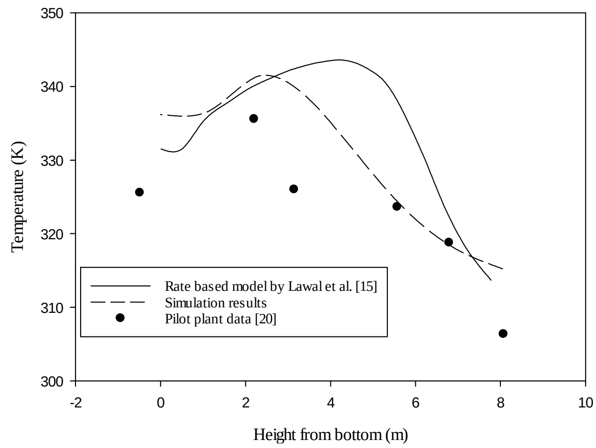

Figure 3: Comparison of absorber temperature profile in dynamic simulation with pilot plant [20] for Case-32 and with simulation results of Rate-based model by [15]

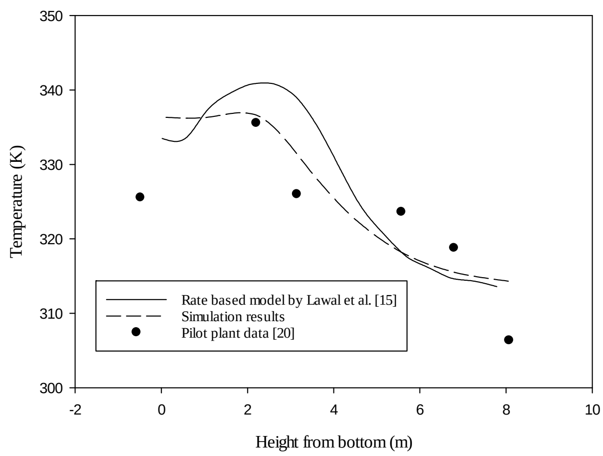

Figure 4: Comparison of absorber temperature profile during dynamic simulation with pilot plant data [20] for Case-32 and with simulation results of Rate-based model by [15] for reduced flue gas flow rate

Therefore, the following inferences may be drawn:

1. An inaccurate mass transfer calculation due to equilibrium models may lead to a prediction of high outlet temperature and rich loading at the bottom of the column.

2. Equilibrium models are relatively accurate for lower liquid to gas ratio operation.

3. Equilibrium-based models for absorbers need to be modified to reduce the higher prediction of mass transfer for operation under high liquid to gas ratio.

3.3 Simulation of stand-alone model of stripper operating in steady state

Similar to the absorber model, the stripper, as shown in Figure 1, was modelled in stand-alone mode with only rich pump, lean pump in the flow-sheet. However, during the building of this flow-sheet at steady state, the lack of available plant data led to an estimation of the thermodynamic conditions of some of the state points. With the stripper specification provided in Table 3 and an operational pressure of 1.6 bar [21], the process flow-sheet for Case 32 was developed and the required state point conditions were estimated. This process flow-sheet was subjected to dynamic simulation with the control systems as mentioned in Table 4. The temperature profile of the stripper was generated and compared, as shown in Figure 5.

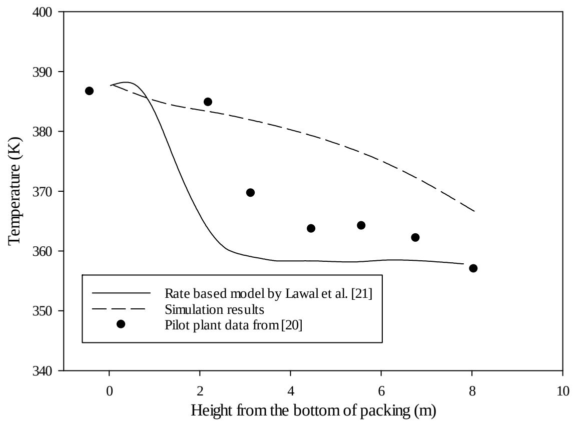

Figure 5: Comparison of stripper temperature profile during dynamic simulation with pilot plant data [20] for Case-32 and with simulation results of Rate-based model by [21]

It may be seen that, with the exception of the temperature for the bottom most point of the stripper, throughout the stripper, the temperatures differ and are higher for the model compared to the pilot plant data as well as for the rate-based model. Due to the control action, the condenser and reboiler temperatures were found to be maintained at the desired conditions; however, an increase in the top most temperature of the stripper led to an increase in condenser duty. This also increased the stripping of CO_2 from the rich MEA stream to some extent and therefore, reduced the lean loading of the plant. As it is known that strippers work very close to equilibrium, the stand-alone models were not able to closely predict the temperature at different stages. The reasons were found to be as follows.

1. Inaccurate prediction of input and output stream conditions.

2. Lower loading at the inlet to the stripper.

In order to avoid such inaccuracies, the complete plant model should be used to estimate the inlet and outlet conditions of the stripper. Improved temperature profile and mass transfer can be observed while estimations were carried out with the complete plant model. This is discussed in the following section.

3.4 Simulation of complete PCC plant operating in steady state

The complete plant, as shown in Figure 1, was subjected to dynamic simulation after building the steady state case based on the pilot plant data for Case 32 and the equipment specification as per Table 3. Missing process conditions in the inlet and outlet of the stripper were estimated in steady state using the complete plant and were used as the initial condition in the dynamic simulation. The temperature profiles for both the absorber and the stripper were generated. These temperature profiles were compared with the pilot plant data and the results of the simulation of the stand-alone models as shown in Figures 6 and 7.

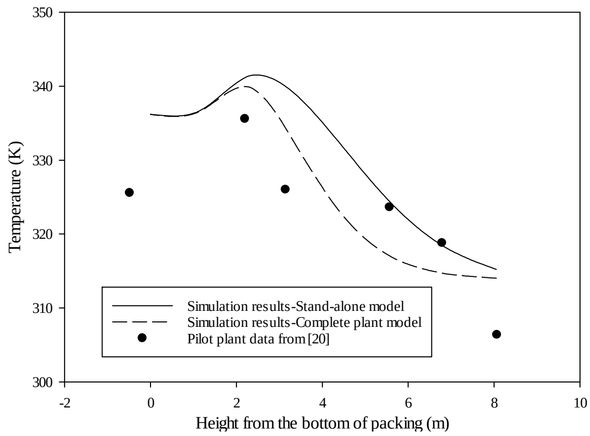

Figure 6: Comparison of absorber temperature profile during dynamic simulation of complete post-combustion plant with pilot plant data [20] for Case-32

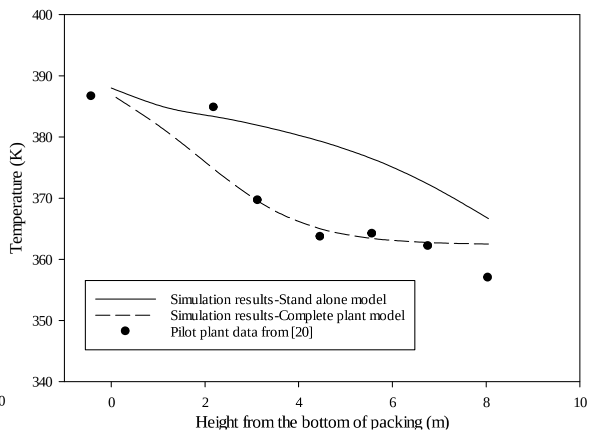

Figure 7: Comparison of stripper temperature profile during dynamic simulation of complete post-combustion plant with pilot plant data [20] for Case-32

The temperature profile for the absorber, as observed from Figure 6, is found to be closer to the pilot plant data, although not exactly the same. The reduction in mass transfer at lower stages of the absorber is the reason for lower loading in the bottom stages of the absorber. This reduction led to lower temperatures at the all the stages in the absorber as compared to the stand-alone model. A reduction in CO2 capture level was also observed from the stand-alone model. The temperature of the stream at the bottom of the absorber was found to be similar to that of the pilot plant.

It may be observed that the stripper temperature profiles for the stand-alone model and the complete plant model are different, with the complete plant model being the more accurate. The temperature at the top of the stripper is also found to be closer to the pilot plant data than that predicted by the stand-alone model. The reason behind the deviation in the stripper temperature profile in the stand-alone model is the incorrect estimation of the state points for the streams in its vicinity, as mentioned in the previous section. The process condition of the stripper mostly depends on the flue gas pressure, absorber temperature and the required lean loading conditions. Therefore, the estimation of stripper conditions should be performed while in a complete cycle; in the earlier case this was performed in stand-alone mode, leading to such high deviation in the performance prediction in the stripper temperature profile. This, thereby, led to a reduction in the condenser duty to below what was previously predicted using the stand-alone model. However, an increase in condenser duty beyond the value of the pilot plant was found inevitable due to the prediction of higher temperature of the top of the stripper as compared to the pilot plant.

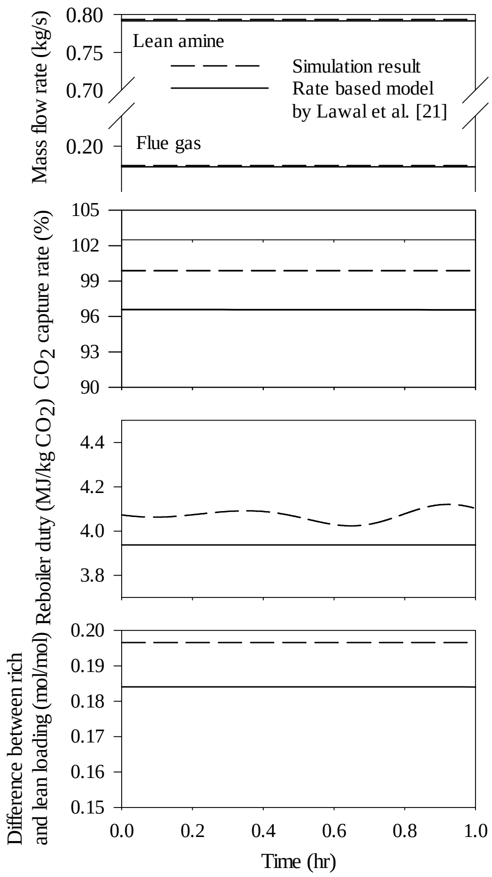

Figure 8: Comparison of simulation results of nominal operation using equilibrium-based model with results presented by Lawal et al., 2010 [21]

Also, a reduction in CO_2 capture from 99.9% to 99.7% was found in the complete plant model; see Figure 8. However, this reduction is negligible compared to the deviation from the pilot plant data. An approximate 5% variation in reboiler duty (3.92 MJ/kg CO_2), as evident from Figure 8, was also found due to proportional variation in rich MEA flow in the complete plant simulation. In addition, this high temperature at the top increased water loss with the CO_2 stream from the condenser. This increased the makeup water flow rate in the plant. However, actual pilot plant data were not found for comparison of all of these variations. Therefore, the dynamic simulation results of [21] were used as a benchmark. Figure 8 presents the comparison of a few important parameters while operating in nominal mode. The following may be noted as observed:

1. Around 3% deviation in prediction of capture rate in the simulation.

2. Reboiler duty prediction also deviates by around 5% as compared to the rate-based results. This deviation is similar to the pilot plant data.

3. The continuous variation of energy requirement in the stripper is due to the variation of liquid level inside the reboiler. A small variation in the reboiler temperature led to a large variation in the heat requirement.

4. The difference between rich and lean loading, however, does not deviate much. The little deviation that can be observed due to inaccurate calculation of rich loading in the absorber.

Therefore, it may be inferred that:

1. Equilibrium models for stripper can be used for dynamic simulation of PCC plants without any modifications in the model; however, reboiler and condenser duty need to be scaled based on the steady state thermodynamic calculation, which would be less than this model predicts.

2. The makeup water flow rate value needs to be recalculated, as this model predicts higher values for it. This is because of the prediction of lower lean loading from the stripper.

3. The heat exchanger has an influence on the operating condition of the stripper. An inaccurate specification or heat transfer coefficient may lead to completely different solution than that expected.

4. A small variation in reboiler temperature led to large changes in heat requirement. However, that small variation did not significantly change the inlet rich MEA stream. Therefore, the control scheme needs to be reevaluated.

3.5 Limitations of use of equilibrium models

It is evident from the above discussions that equilibrium-based models for the absorber predicts higher than actual mass transfer. This is more apparent for processes with higher liquid to gas ratio. As earlier researchers have mentioned, absorbers work far away from equilibrium and dominated by reaction kinetics, simple equilibrium-based models should not be used for simulation. In the case of stripper, such models have been found to be sufficient as strippers work close to equilibrium conditions. However, from above discussions, the level of inaccuracy in the simulation results while using such models was discovered as follows:

1. Calculated reboiler duty may deviate in a range of up to 5% from the actual.

2. The calculation of CO_2 capture rate may deviate between 3-12%.

3. Prediction of rich loading may deviate within 1% from the actual.

4. Overall mass balance may have a deviation much less than 1%.

Therefore, the major limitation of using such models is the high deviation in mass transfer calculation leading to the prediction of an unacceptably high CO_2 capture rate, which directly affects the reboiler duty in the stripper and the rich loading. This requires suitable modifications to the simple models used in the simulator. As stated in the literature, the alternative to this is to use rate-based models. However, the possible scope for modifying this equilibrium models was investigated and is presented in the following section.

4. Method for improving of accuracy of performance prediction using equilibrium-based models

In order to justify improving the equilibrium-based models rather than directly using the rate-based ones, it is important to look back at the purposes of dynamic simulation as they dictates the use of rigorous models providing the highest accuracy. In general, the purposes of dynamic simulation in PCC processes are as follows.

1. To understand and analyze the behavior of these plants with designed control structures during part load operations, to operate flexibly with power plants etc., and to identify the process and control modifications require for the optimum performance of the processes,

2. Using in self-tuning controllers or using with advanced controllers such as model-based predictive controller etc.,

3. To investigate the process response during start-up and shutdown and optimize the control structure for desired procedure for such transients,

4. To evaluate the performance of large equipment during transients and identifying possible cause effect matrix for tripping the operation of plants due to some specific transient operations,

5. To determine the equipment failure and safety,

6. To identify both the environmental impact of transients in the plants and suitable controls to minimization of the effects on the environment,

7. Operator training and assistance during operating plants, etc.

In most cases, highest order of accuracy in simulation results is not mandatory; in fact, simpler models with known inaccuracies are acceptable [16]. Therefore, researchers have proposed the simplification of rate-based models. However, the scope for improving the equilibrium models needs to be evaluated as it is easier to include equations in a simpler model than to reduce from the complex and intertwined set of equations. An attempt was made to improve the equilibrium models as discussed in the following section.

4.1 Method to improve accuracy of simulation results using equilibrium models for absorber

As mentioned in previous section, equilibrium models for absorbers are the most inaccurate in terms of calculation of mass transfer rate. It is known that, ideally, absorbers work far away from equilibrium. Therefore, in steady state simulation Murphree efficiencies have been used in several studies [22, 25, 26]. These take component fractions into consideration while calculating the stage efficiency for individual component. In a way, this constraints the component fractions at desired values when the packing is divided into theoretical stages. However, it is not possible to include it in dynamic simulation using the process simulator and also, as the holdup in the stages varies with the change in flow rate, constraining the component fraction may lead to inaccurate calculation of heat and mass transfer. The two-film theory, which is widely accepted as appropriate for interphase mass transfer calculations, considers mass transfer at the interface of liquid and vapor. According to this theory, instantaneous diffusion between liquid and vapor leads to equilibrium at the interface if the reactions are fast. However, bulk fluid remains homogeneous and well mixed [16]. Therefore, it may be considered that a small portion of both liquid and vapor remains in equilibrium where heat and mass transfer occur.

In the process simulator, there is an option to bypass the desired fraction of the vapor mass flow rate from the individual stages [27]. This is considered as stage efficiency. This can be useful as bypassing part of vapor from interacting with the liquid may help to reduce the overall mass and heat transfer. Also, mixing the bypass flow with the vapor coming out of any stages can lead to the desired temperature profile throughout the absorber. It is due to increase in heat transfer as the inlet vapor temperature to the stage above reduces and thereby increases the temperature difference in the liquid vapor interface. Therefore, a parametric study was performed to identify the optimum stage efficiency; this was found to be 0.4 for the absorber, as can be observed in Table 7. This was specified in the dynamic model of the absorber by allowing the lower vapor to be in contact with the liquid in the individual stages in order to improve the accuracy of calculation of mass transfer in the stages. However, the stripper was allowed to work in equilibrium mode as its use was found to be acceptably accurate. The results of simulation using reduced stage efficiency were compared with the earlier results and the results of the rate-based model as listed in Table 7.

Table 7: Comparison of normal operation simulations using with and without reduced stage efficiency and with rate-based model by Lawal et al., 2010 [21]

Parameter	**Simulation without reduced stage efficiency**	**Simulation with reduced stage efficiency of 0.35**	**Simulation with reduced stage efficiency of 0.4**	**Simulation with reduced stage efficiency of 0.45**	**Rate-based model by Lawal et al., 2010**
CO_2 capture rate (%)	99.87	93.82	96.62	97.55	96.59
Reboiler duty (MJ/kg of CO_2)	4.07	4.18	4.15	4.13	3.94
Difference between rich and lean loading (mol/mol)	0.197	0.187	0.187	0.189	0.184

A significant improvement was observed in the CO_2 capture rate after the addition of the stage efficiency. This rate was found to be similar to that predicted by the rate-based model. It was also observed that the rich loading from the absorber was reduced, leading to a decrease in the difference between rich and lean loading, close to that predicted by the rate-based model. However, in order to maintain similar lean loading, the reboiler duty was found to be increased even above the earlier estimation. This improvement was identified with the steady state normal operation simulation using the dynamic model with a constant stage efficiency values of 0.4. Further, the use of this factor as reduced stage efficiency in the absorber during transients in the process needs to be verified. Therefore, the variation in flue gas flow rate was simulated by maintaining the liquid to gas ratio in the process. The simulation results are compared with the same data presented in the literature while using rate-based model [21]. A ramp increase of 10% in the flue gas flow rate was applied over a period of 10 min and variation in capture rate and reboiler duty were recorded and compared with the data obtained from literature. A similar variation in lean MEA flow by controlling liquid to gas ratio was obtained in the simulation, as evident from Figure 9.

During the change in flue gas flow rate to a new steady operating condition, with our model, the prediction of settling time for the capture rate was found low, compared to the simulation results obtained from literature. Therefore, this model predicted a minimum capture rate of 96.6% compared to 96.3% using rate-based model and the process was found to reach within ±1% variation of output by 15 mins after application of the change in flue gas flow-rate. This value is much less than that predicted by using rate-based model. The reasons may be as follows:

1. Inaccurate estimation of thermal inertial involved in the process, e.g. in the heat exchanger. This thereby led to a reduction in the settling time of outputs.

2. Ignoring the piping and corresponding thermal mass may have influenced the rate of change in the temperature at the inlet of the stripper.

3. Ignoring the storage of rich MEA after coming out of the absorber led to an instantaneous change in inlet temperature and rich loading to the stripper. However, with absorber sump this variation cannot be instantaneous. This led to the prediction of a lower settling time.

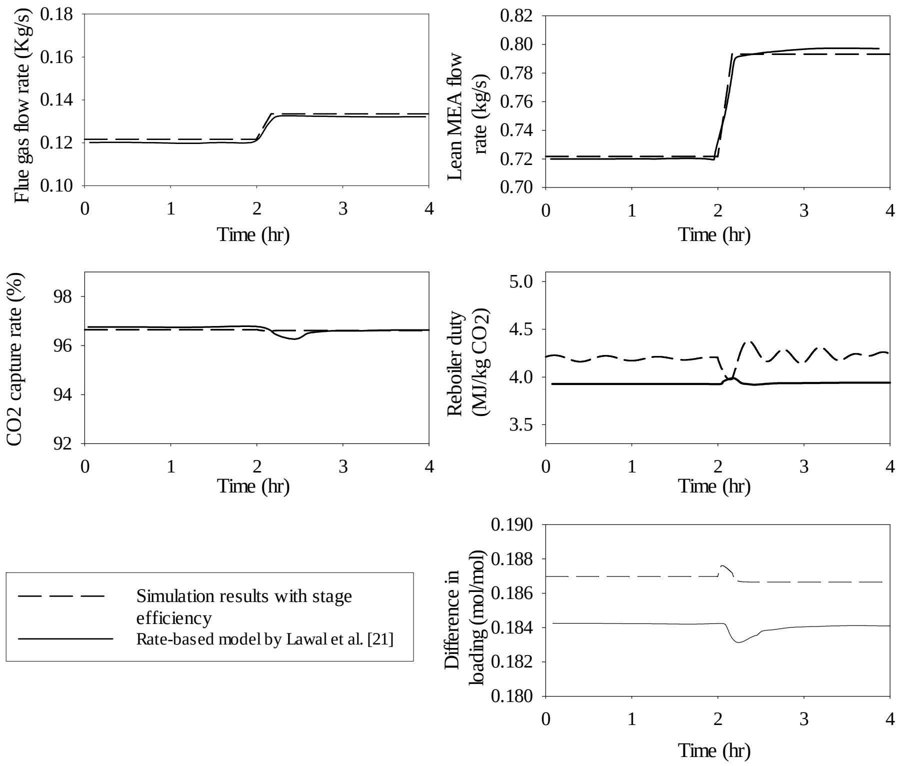

Figure 9: Variation of important parameters while applying step change in flue gas flow rate as compared to the simulation results using rate-based model by Lawal et al., 2010 [21]

All of these have influenced a change in rich loading in the cycle to the corresponding condition, which is faster compared to the results of the rate-based model. Thereby, this increased the rate of change in the capture rate to above what it should be, as shown by the results of the rate-based model. The higher rate of change in rich loading also caused the specific reboiler duty to reduce as the heat duty did not change significantly as the CO_2 capture increased. However, the specific reboiler duty increased as soon as the heat duty reached the required value corresponding to rich loading, see Figure 9. Also, as the lean MEA has storage as reboiler, the lean loading did not change as fast as the rich loading. Therefore, the difference between them has increased instead of decreasing. Therefore, when considering transients, the piping and absorber sump are important equipment, as they are mostly capacitive components and mainly affect the settling time of the process parameters due to the higher thermal inertia. However, the inclusion of these equipment in the process is outside the scope of this study. In future, work needs to be done to incorporate all of these equipment and, in doing so, to include their effects in process transient of the cycle.

4.2 Pros and cons of the method

It was observed that the inclusion of stage efficiency greatly improved the performance prediction of the PCC process using the equilibrium-based model. Among others, the capture rate, performance of the controller for maintaining the liquid to gas ratio, the difference of rich and lean loading in cycle were found to be similar to the pilot plant data and the simulation results of the rate-based model. A maximum deviation of around 5% was also obtained for the reboiler duty using the similar modified model. Therefore, this method of reduced stage efficiency in equivalent stages in the absorber with equilibrium models can be one of the alternatives to using complex rate-based models. The major benefit obtained by using this is an improvement in the time required to simulate PCC processes in dynamics. It is noteworthy that this cannot be considered as a replacement for rate-based models; nevertheless, continuous modification of this model can further improve the simulation results. However, the following issues need to be addressed:

1. The theoretical basis of this method needs to be identified.

2. Stage efficiency is envisaged to be a function of packing type; therefore, for different kinds of packing it is necessary to find out the fraction of vapor to be bypassed.

3. It is necessary to discover whether the stage efficiency remains constant or varies within a maximum range with a change in absorber dimensions and packing type.

Therefore, it can be stated that the approach of modifying the equilibrium-based models to improve the accuracy of simulation results can be an alternative to the approach of simplifying complex rate-based models, for use where simulation time is more important than obtaining exact solutions. Other ways to improve such equilibrium-based models also needs to be investigated.

5. Transient response of pilot plant under variable process conditions

When integrated with power plants, PCC plants are required to cope with the operational requirements of power plants. It has been estimated that there will be a demand for operational flexibility in futuristic power plants due to the high penetration of renewable energy sources in the European energy market by the year 2030 [28]. Therefore, one of the major concerns for the operation of PCC integrated with such power plants is how much these comparatively slow and semi-batch chemical processes can cope with during such flexible operation of power plants. Researchers have used dynamic simulation to address this problem. PCC plant behavior, with variations in flue gas flow rate, steam extraction from the power plant, reboiler duty, etc., has been analyzed [21, 24, 29, 30, 34]. Different techniques such as storage of rich/lean MEA, bypassing exhaust gas, etc., have been evaluated using dynamic simulation to enable PCC plants to operate stably with high power plant load fluctuations while maintaining the required CO_2 capture rate, either according to time-averaged value or throughout the operation [16]. Dynamic simulations have also been used for designing suitable control structures using plantwide control methods [31-33]. However, few studies have compared the effects of controlling the liquid to gas ratio and the CO_2 capture rate when the PCC plants have to operate under fluctuating flue gas conditions without using any techniques to mitigate those fluctuations. Here, an attempt was made to analyze the same, using a hypothetical scenario of a combined cycle gas turbine (CCGT) power plant.

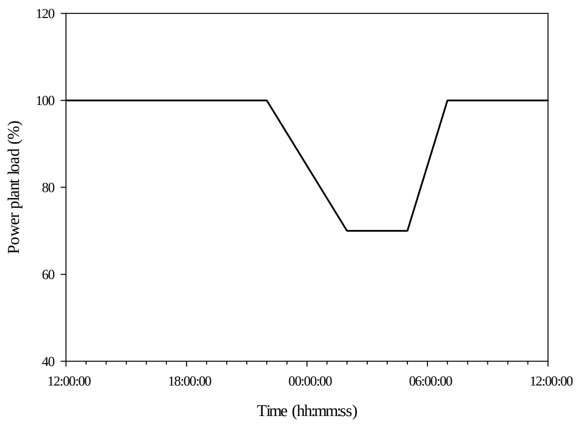

Figure 10: Hypothetical power plant load variation during a 24-hr operation adopted from [16]

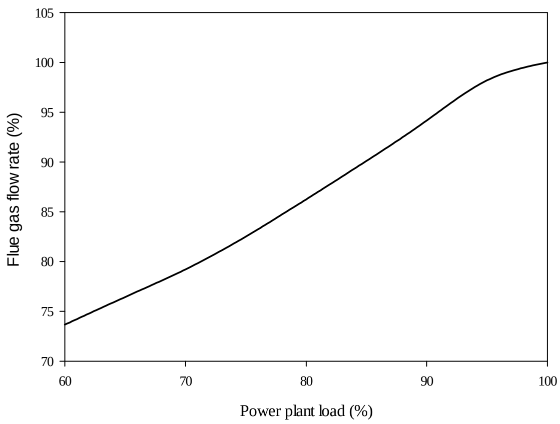

Figure 11: Typical flue gas flow rate variation during part-load operation of power plants

The hypothetical scenario in variation of daily power plant load, depending on the electricity price and demand, as presented by [16], was considered here to analyze the performance of the pilot plant using the dynamic model; see Figure 10. This typically represents the variation in power plant load throughout the day, starting at 12.00 hr of any given day of the year. The corresponding flue gas flow rate variation can be obtained by typically scaling the large-scale power plant output. It can be found that variation in flue gas flow rate is not proportional to that of power plant load changes. A typical CCGT power plant exhibits flue gas flow rate variation with the change in plant load conditions, as shown in Figure 11. The mass flow rate for the pilot plant is scaled according to this characteristic and subjected to the plant in dynamic simulation. It is noteworthy that with the change in power plant load, the air and fuel flow-rate to the gas turbine changes and correspondingly, the mole fraction of CO_2 also changes. However, in the current study, this variation was not considered. As previously discussed, the two following scenarios were considered:

1. Controlling liquid to gas ratio (L/G ratio),

2. Controlling CO_2 capture ratio.

The 24 hours of operation were simulated with these two different control structure. The results of the simulations are presented in Figure 12. It can be seen that for except once in the case of lean MEA flow rate for controlling the *L/G* ratio, discontinuity has not been observed in the simulation.

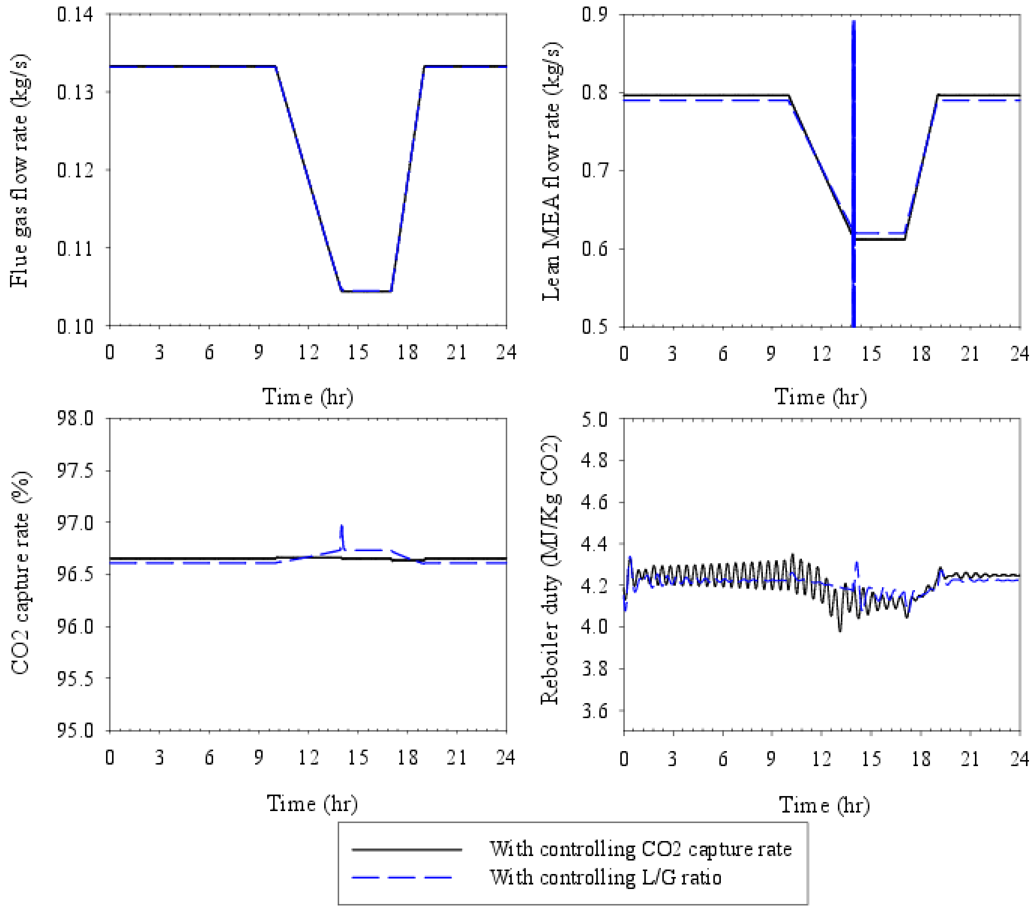

Figure 12: Simulation results of plant behavior corresponding to variable power plant load condition

It is evident from the results that with both control structures, the process worked stably with constant separation of CO_2 from rich MEA, providing a mole fraction of 93.2% in the exhaust from the stripper and continuous variation in the reboiler duty within a small range of less than ±1%. However, the CO_2 capture rate varied from 96.6% to as high as 96.7% with the time-averaged capture rate being less than 96.65% when the *L/G* ratio was controlled. Therefore, set point tracking needs to be included for maintaining time the average capture rate at the desired value. On the other hand, controlling the capture ratio was found to maintain the time-averaged capture rate at almost 96.65% for the 24 hours of operation. However, continuous variation in liquid level in the reboiler led to a variation in the heat duty in the reboiler. These variations in turn require changes in steam extraction from the power plant. However, in CCGT power plants it is not desirable to impose continuous fluctuations in steam extraction. This requires analysis of the bottoming cycle of the power plant, i.e. the steam cycle, to provide such flexibility. Using the *L/G* ratio controller eliminated the requirement for such fluctuations in heat duty and thereby provided much stability in steam extractions. Also, the separated CO_2 flow rate with consistent CO_2 mole fraction of 93.2% varies from 0.365 kg/s to 0.286 kg/s from the stripper. With the continuous change in the reboiler duty when the capture rate was controlled, almost ±3.5% fluctuation in the CO_2 flow rate was also observed, which was absent in the case of controlling the *L/G* ratio.

Based on the above mentioned observations, the following advantages and disadvantages of the use of both controllers were found.

	Controlling capture rate	**Controlling L/G ratio**
Advantages	1. Helps in maintaining desired time average capture rate. 2. Low lean MEA requirement during low power plant load conditions. 3. Reduced reboiler duty required during operation under flexible power plant operation over a period of 24 hours. 4. Lower settling time for the output compared to the other controller.	1. Helps in maintaining L/G ratio throughout. 2. Helps in stabilizing the liquid level in reboiler and thereby minimizing fluctuations in reboiler duty. 3. Results in lower fluctuations in CO_2 product from stripper leading to stable upstream operations.
Disadvantages	1. Leads to fluctuations in reboiler liquid level resulting in continuous variation in both reboiler duty and CO_2 product from stripper. 2. Higher lean MEA requirement during full load condition of power plant.	1. Unable to response towards maintaining desired time-averaged capture rate. 2. Higher reboiler duty required during operation under part load condition of power plant.

Therefore, it can be inferred that controlling the capture rate during the operation of the power plant under a flexible operational scenario can be chosen over controlling the *L/G* ratio where stringent emission requirements are the objective. As the mole fraction of CO_2 in the flue gas flow varies during the part load operation of the power plant, it is important to further analyze both the controllers in respect of identifying their suitability. In future, efforts need to be made to study this.

6. Conclusions

In this paper, the limitations of using equilibrium-based models for simulating post-combustion CO_2 capture processes were identified. Simple equilibrium-based models for the absorber may lead to as high as 12% deviation in the prediction of the CO_2 capture rate, compared to pilot plant data. However, equilibrium-based models for strippers may be used with an accuracy penalty as high as 5% in the prediction of reboiler duty. The method using reduced stage efficiency in the absorber to improve the simulation results has been demonstrated in the literature. However, a constant value for the stage efficiencies was found to be used for any packing types. Parametric analysis in this paper revealed that this should be adjusted based on the packing type and identified stage efficiencies were found to differ from the values mentioned in the literature. This method, with identified reduced stage efficiencies in the absorber, was found to be suitable, and, using this method, reasonable accuracy was obtained in dynamic simulation results. A maximum deviation of 5% for the calculation of rich loading was found in the simulation results. While instantaneous response of the plant was found to differ from that predicted by the standard rate-based models, the reasons for such deviations were identified; these issues need to be addressed in future work. It is noteworthy that modification of such models did not affect the time required for simulations. Therefore, it can be concluded that the approach of modifying equilibrium-based models for PCC processes to improve accuracy can be an alternative to the approach of simplifying rate-based models for simulations where the required simulation-time is more important than obtaining exact solutions.

The modified equilibrium-based model was later used for the simulation of the transient condition of a ramp change in the flue gas flow rate. The flue gas was varied according to the hypothetical characteristics of future power plants. With the objective of comparing two control structures – controlling liquid to gas ratio and controlling the CO_2 capture rate – separate models were developed and analyzed. It was observed that with the liquid to gas ratio controller it was difficult to maintain the time-averaged capture rate, despite keeping the composition of both streams similar. Therefore, the CO_2 capture rate need to be controlled explicitly as while the power plant would be operating under part-load conditions, there would be as high as a 10% variation in CO_2 content in the flue gas flow rate. With the liquid to gas ratio control, this variation could not be taken care of and that might have led to a lower capture rate. However, it will be interesting to investigate the performance of such controls with variation in the mole-fraction of CO_2 in the flue gas. Also, the following issues needs to be addressed in future work.

1. Identification of the relationship between packing type and stage efficiency for absorbers. For this purpose, different available pilot plant data can be used and need to be compared while using this dynamic model.

2. Inclusion of absorber sump and piping in the dynamic model of PCC processes. This will change the performance prediction of the processes by incorporating the dead-time in the overall settling time of the process output during transients.

3. Identification of the parameter to measure for stably controlling the reboiler temperature. It is important to stabilize the steam extraction from the power plant during part-load operation.

4. Analysis of the control structure for investigating the effects of variation of CO_2 mole-fraction in the flue gas flow-rate and also the reason behind difficulties in rejecting various disturbances in the PCC plants during operation under periodic flue gas change.

Acknowledgements

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