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Applicability and validation of use of equilibrium-based absorber models with reduced stage efficiency for dynamic simulation of post-combustion CO_2 capture processes

Rohan Dutta[a]*, Lars O. Nord[a], Olav Bolland[a]

[1]*Department of Energy and Process Engineering, NTNU-Norwegian University of Science and Technology, Trondheim, Norway*

Abstract

Use of complex rate-based models for absorbers in a post-combustion CO_2 capture process may lead to high computational complexity and simulation time. Method using reduced stage efficiency in equilibrium-based model for such columns may provide acceptable range of accuracy with comparatively much less simulation time and thereby enabling them to be used simultaneously with dynamic models of power plants. This paper discusses the applicability of such method and presents the validation of use of such methods using two pilot plant data at both steady state and transient operation. Steady state plant data were used to determine the reduced stage efficiencies for the pilot plants. The dynamic models with those stage efficiencies were simulated to predict both the steady state operation and transient operation under step change in solvent flow rate scenario with nominal deviations in a range of 3-6% in the simulation results with simulation time 35 times higher than the real-time.

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Keywords: Post-combustion CO_2 capture; Unisim Design; Pilot plant; Absorber modelling

1. Introduction

Dynamic modelling and simulation of post-combustion solvent based CO_2 capture (PCC) plants is of interest for investigation of their operability and for identifying suitable control structures while integrated with fossil-fuel based

* Corresponding author. Tel.: +47-9808-3693.
E-mail address: rohankrdutta@gmail.com

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power plants [1]. Additionally, the plant's dynamic model can be used in advanced controllers such as model predictive controllers etc. Dynamic models for such purposes require, on one hand, a high fidelity and robustness and, on the other hand, low computational time.

Two major equipment of PCC processes, absorbers and strippers, may be modelled using the two approaches of equilibrium-based and rate-based modelling. It has been reported that use of rate-based models with reaction kinetics provides higher accuracy in simulation results compared to equilibrium-based models. However, the simulation time has been found to be high and thereby researchers have suggested simplification of such models for the purpose of, e.g., model predictive control [2, 3]. On the other hand, equilibrium-based models with reaction kinetics provide simpler dynamic models for both absorber and stripper leading to lower computational time [4]. Use of such models reduces accuracy of prediction of heat and mass transfer and thereby produces higher capture rate, rich loading, and reboiler duty. In addition, it has been observed that the absorber temperature profiles produced by such models highly deviate from temperature profiles obtained from pilot plants [2].

In building the rate-based models, the two-film theory has mostly been used [2, 3, 5-10]. The basis of the two-film theory is gas-liquid interface mass and heat transfer and equilibrium has been considered at the interface. It reduces the equilibrium interaction of gas and liquid. Therefore, limiting gas from entering each stage of an equilibrium-based model for absorber can help in reducing the equilibrium interaction of liquid and gas and thereby can decrease heat and mass transfer at each stage. However, this may lead to different stage temperatures in the column and the range of deviation, however, need to be identified. This option of bypassing a part of the gas from entering the equilibrium stages can be implemented using the process simulator UniSim Design R430 by specifying stage efficiency (y) as shown in Figure 1. Stage efficiency is the fraction of gas bypassed from each stage. The stage efficiency is comparable to multicomponent Murphree vapor phase efficiencies, however, different in values those further reduce the interaction of liquid-vapor.

In this paper, attempt was made to validate the applicability of use of reduced stage efficiency for the equilibrium-based absorber models for using them along with the dynamic models of power plants in order to study the operational flexibility of such integrated plants. For the purpose of validation, both steady state and transient operational data of one large pilot plant and another small pilot plant were used [2]. Therefore, the objective of this paper is as following.

1. Determination of stage efficiency of absorber models for two pilot plants using the steady state operational data.
2. Verification and validation of simulation results using the determined stage efficiencies using steady state and transient operational data.

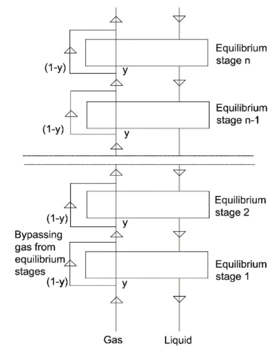

Fig. 1. Bypassing gas from equilibrium stages of absorber.

2. Methodology

2.1. Configurations of the pilot plants

Operational data for two pilot plants at Gløshaugen and at Tiller were obtained from [2]. The process flow-sheets for both the plants are similar or derivatives of base case, shown in Figure 2, with different equipment sizes and process conditions. Pilot plant process conditions and equipment specifications are shown in Table 1 and 2, respectively. Following assumptions were made while preparing the cases for both the pilot plants.

1. No piping was considered. Pressure drops and residence time were included, wherever applicable, in the neighboring equipment such as heat exchangers and valves.
2. Absorber sumps were considered both in steady state and dynamic models.
3. Constant lean amine flow-rates and lean loading were assumed at the inlet of the absorbers unless otherwise stated.
4. Constant CO_2 mole fraction in the flue gas was considered unless otherwise stated in the dynamic simulation.
5. Make-up streams and mixing of CO_2 desorbed with treated gas were ignored in dynamic simulation.
6. Heat loss was ignored and overall heat transfer coefficient in the heat exchangers were consider constant instead of flow dependent.

Table 1. Pilot plant process conditions [2].

Stream	Parameters	Gløshaugen pilot plant		Tiller pilot plant
		Case 1	Case 2	
Flue gas	Flow rate (Nm^3/hr)	78.6	75.8	216.4
	CO_2 composition (vol%-dry)	3.52	6.77	4.2
	Temperature (^{0}C)	42.2	49.2	48.4
Lean amine	Flow rate (kg/hr)	166.9	172.3	222
	Amine wt-%	30	30	27.1
	Temperature (^{0}C)	49.1	58.3	41.6
	Lean loading (mol CO_2/mol MEA)	0.3	0.26	0.14
Rich amine	Rich loading (mol CO_2/mol MEA)	0.42	0.45	0.48
Treated gas	CO_2 composition (vol%-dry)	0.29	1.19	-

Table 2. Specifications of equipment of both the pilot plants used in the paper [2].

Equipment	Parameter	Gløshaugen pilot plant	Tiller pilot plant
Absorber	Diameter (m)	0.15	0.20
	Height (m)	4.23	19.42
	Packing type	Sulzer BX	Sulzer Mellapak 2X
Regenerator	Diameter (m)	0.10	0.15
	Height (m)	3.57	13.78
	Packing type	Sulzer BX	Sulzer Mellapak 2X
Lean/Rich heat exchanger	Overall heat transfer coefficient (MJ/C-h)	11.6	10
	Temperature approach (^{0}C)	5	5
Condenser	Temperature (^{0}C)	15	-
Reboiler	Temperature (^{0}C)	117	117.4
	Residence time (mins.)	14.4	40

2.2. Process simulator

The pilot plants were first simulated at steady state using rate-based column models in Aspen HYSYS. These rate-based models utilizes the RateSep subroutine for determination of component efficiency for each stages based on the tray/packing specifications. Based on the plant data available, the steady state cases were constructed and results were also compared with that of pilot plant. Dynamic models in UniSim Design were built on the steady state results as obtained from HYSYS.

The dynamic column models in UniSim Design are based on equilibrium-based, which utilizes the principle of equilibrium condition at each stages. Corresponding stage mass, energy and component balances, and stage hold-up used for modeling the columns. Amine property package in the simulator was used with Kent and Eisenberg model for equilibrium solubility and Peng-Robinson equation of states for calculating the fugacity coefficients.

The stage efficiency in the column models is defined as the ratio between the gas bypassing from entering the equilibrium stage and total gas flow-rate to that specific stage, see Figure 1. This allows constraining the gas and liquid interaction that in the present simulator is considered at equilibrium condition. In this paper, this method is used for reducing the liquid to gas equilibrium interaction in order to replicate the interface mass transfer predicts by the rate-based models. It is to be noted that this stage efficiency is an overall value unlike the stage efficiencies that are used at steady state simulation for each acid gas components.

Absorbers of both the pilot plants were modeled using above mentioned equilibrium-based column models with reduced stage efficiencies. However, simple equilibrium models for the regenerators/desorbers were used in this study as it has been previously reported that these columns worked close to equilibrium and they could be modeled accurately using simple equilibrium models [2, 4].

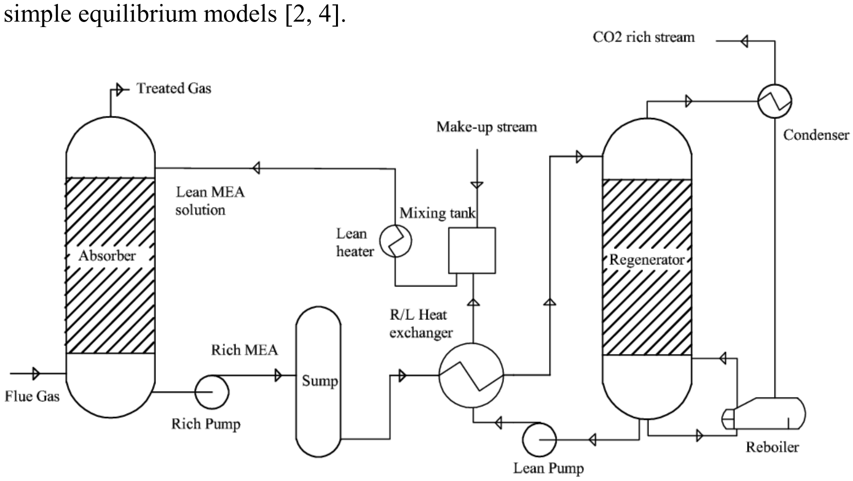

Fig. 2. Basic process flow-sheet of pilot plants.

2.3. Solution methodology

Euler implicit method was used for solving the algebraic-differential equations with a time step size of 0.5 s. Results of simulation for selected parameters were recorded after every 20 s using the data logger in the simulator. 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 streams, outlet streams and vessel wall of all equipment at atmospheric conditions. Pressure or flow specifications in boundary streams of the flow-sheet and flow specifications at the intermediate streams in the regenerator were used. Solutions obtained from steady state simulations for streams at steady state conditions were used as the initial values during the dynamic simulations.

3. Determination of stage efficiencies for dynamic models

Both the pilot plants use different structured packing in absorbers. As in packed columns, the surface area of gas to liquid interaction is provided by the packing material via parameters such as packing factor, void fraction and specific surface area, the stage efficiency may be required to be determined every time as the packing material changes and the values can be different. It is because of this reason, operating data of two different pilot plants were used. Parametric studies were performed to determine stage efficiencies those led to minimum deviations in simulation results compared to the steady state operating data from the pilot plants. However, it requires to be verified that with the change in process operating conditions the stage efficiency remains same. Therefore, upon determined the stage efficiency for the pilot plant columns, validation was performed with different process conditions than that used for determination.

3.1. Based on the steady state operational data of Gløshaugen pilot plant

As stated in earlier sections, the dynamic models of both the pilot plants in UniSim Design were built based on the steady state simulation results obtained from modeling in HYSYS. This models were fully specified before moving to the dynamics. Typical control systems used in the pilot plants were included in the flowsheet and they were tuned using the steady state operating conditions. In most of the controllers, PI-type of controllers were used, however, except for the controllers for reboiler level, which were controlled using P-only controller to avoid any unwanted oscillation due to change in operating conditions.

The stage efficiency can be varied in a range of 0 to 1. For both the pilot plants, simulations were started from 1 and with 0.1 step stage efficiencies were changed and required data were recorded. The parameter, difference in CO_2 capture rate, was considered to be the parameter to be minimized by changing the stage efficiencies in the absorbers as this dictates most of the process parameters such as rich loading, flow rate, reboiler duty as well as lean loading. Therefore, minimizing it would eventually lead to optimizing other parameters.

The model of Gløshaugen pilot plant was simulated under the operating conditions for case 1, see Table 1. After specifying the stage efficiencies in the model, the flowsheet was simulated until the simulation reached steady state conditions similar to pilot plant and upon reaching there the capture rates and CO_2 mole fraction in the treated gas were noted. Differences of obtained capture rate in the simulation and that of the pilot plant were determined. Results are shown Figure 3 and 4. It can be seen that the minimum difference in the capture rate from pilot plant data can be obtained for stage efficiencies of 0.4. It is also evident from the variation of difference in CO_2 mole fraction at treated gas between simulation and pilot plant that with minimum difference in capture rate, the difference of it also is minimum. This justifies our selection of parameter for identifying the stage efficiencies.

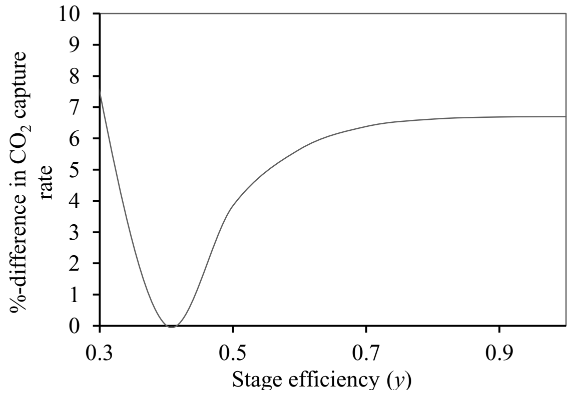

Fig. 3. Variation of difference of CO_2 capture rate with changing stage efficiency (y) in the absorber.

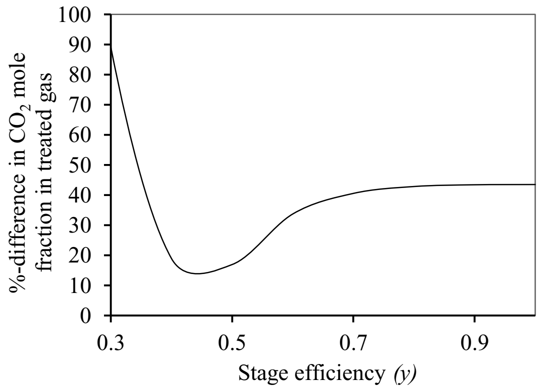

Fig. 4. Variation of difference of CO_2 mole fraction in the treated gas with changing stage efficiency (y) in the absorber.

3.2. Based on the steady state operational data of Tiller pilot plant

Similar to the method used to identify the stage efficiencies for Gløshaugen pilot plant, the dynamic model of Tiller pilot plant was simulated and variation of difference in capture rate was determined. The results are shown in Figure 5. It can be observed that the minimum difference in capture rate was obtained at stage efficiencies of 0.8. It is different and higher than that obtained for the other plant. It is also observed that for the packing used in this pilot plant worked closely to equilibrium conditions with high interface area for liquid and gas. Less than 0.02% deviation was observed with complete equilibrium model with stage efficiencies equal to 1. Therefore, use of reduced stage efficiencies in this case may be avoided if required. The trend in changing the difference in capture rate is found to be similar to that of the other pilot plant.

It is noteworthy to mention here, that both the pilot plants are having almost similar absorber diameter, however the height of the absorber in the Tiller plant is much higher than the Gløshaugen pilot plant. This eventually leads to higher stage height, stage volume and the overall pressure drop in the absorber at Gløshaugen pilot plant compared to the other plant. As it was observed that the absorber at Tiller plant was found to be working close to equilibrium, it will be interesting to study if such designs helps in achieving operation close to equilibrium conditions, i.e. increasing liquid and gas interface and thereby increasing the interaction between them.

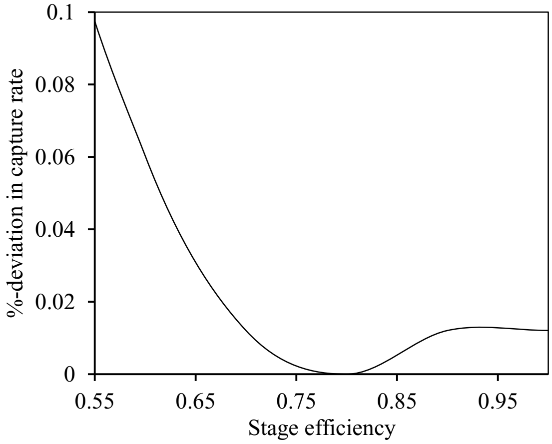

Fig. 5. Variation of difference of CO_2 capture rate with changing stage efficiency (y) in the absorber.

Identified stage efficiencies were used for simulation of both the plant models under different operating conditions and validated the simulation results using both steady state and transient operational data of the pilot plants as discussed in following sections.

4. Validation of dynamic simulation of pilot plants

The stage efficiencies identified are 0.4 and 0.8 for Gløshaugen and Tiller pilot plants, respectively. This values were specified in the absorbers and the scenario of steady state and transient conditions as of case 2 for Gløshaugen plant and the steady state operation of the Tiller plant were simulated. The results were compared with the available plant data for absorber, desorber temperature profiles, absorption and desorption rates, rich, lean loading etc.

4.1. Using steady state operational data

The operation as case 2 in Gløshaugen pilot plant was performed with higher CO_2 content in the flue gas. This led to increase lean amine flow rate and correspondingly higher reboiler duty than the case 1. Both the cases were simulated again with stage efficiencies of 0.4 and important parameters were compared with the plant data. The results of simulation are shown in Table 3 and Figure 6 and 7. A comparison with the steady state simulation in HYSYS has also been shown to confirm the deviation of the steady state simulation results from the pilot plant. It can be observed that they are close to the plant data and therefore can be used as reference for the instances when pilot plant or existing plant data are not available.

It can be observed that simulation predicted less than 2% deviation in capture rate than plant data for both the cases and almost similar to what were calculated using HYSYS at steady state simulation. Although, reboiler duties for both the cases were estimated higher in dynamic simulation and the temperature profiles of both absorber and desorbers for case 2 differ from the plant data. The reason behind these are:

1. In the simulation equilibrium reactions were considered. Improved temperature profiles can be obtained using reaction kinetics.
2. Lower heat exchanger overall heat transfer coefficient was estimated. Also variation of it with flow rate changes need to be included to increase the accuracy of prediction of inlet temperature to the desorber. This can lead to reduction in reboiler duty and improve the prediction of the model.

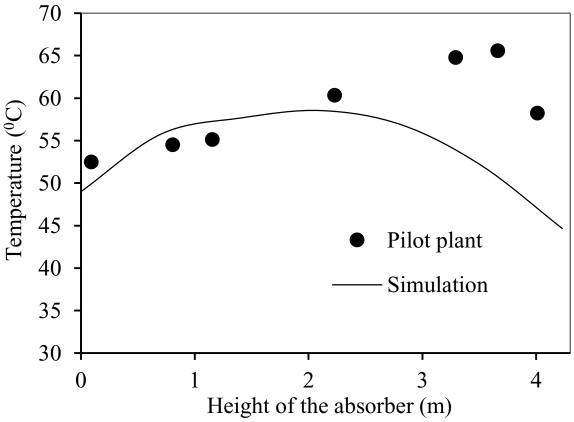

Fig. 6. Absorber temperature profile for case 2 with 0.4 as stage efficiency (y) in the absorber Gløshaugen pilot plant.

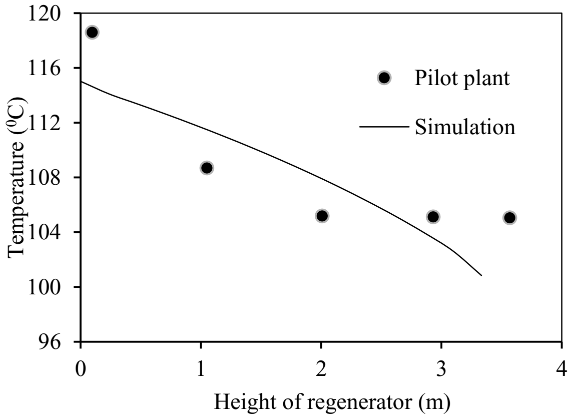

Fig. 7. Regenerator temperature profile for case 2 with 0.4 as stage efficiency (y) in the absorber in Gløshaugen pilot plant.

Table 3. Comparative results of simulation of Gløshaugen pilot plant (SS: steady state simulation, Dyna: dynamic simulation).

Parameter	Pilot plant data [2]		Using rate based model at SS		Using equilibrium based model at Dyna (y=0.4)	
	Case 1	Case 2	Case 1	Case 2	Case 1	Case 2
Lean loading (mol CO_2/mol MEA)	0.30	0.26	0.30	0.26	0.30	0.26
CO_2 capture rate (%)	88.69	83.46	86.91	80.21	88.70	85.40
Rich loading (mol CO_2/mol MEA)	0.42	0.45	0.39	0.44	0.40	0.45
Reboiler duty (kW)	6.50	9.23	6.77	9.45	6.79	10.00

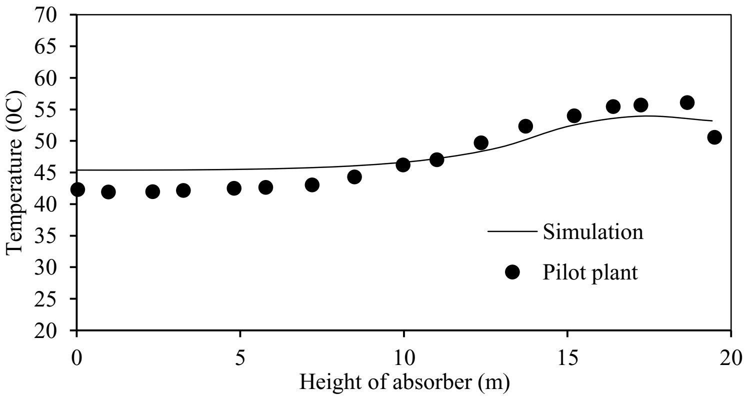

Fig. 8. Absorber temperature profile with 0.8 as stage efficiency in the absorber in Tiller pilot plant.

In contrast to the Gløshaugen plant model, the model of Tiller plant with reduced stage efficiencies of 0.8 was found to be predicted similar temperature profile in the absorber as compared to the plant data, see Figure 8. Also, it can be observed from Table 4 that parameters such as captured CO_2 from gas (<3%), desorbed CO_2 (<6%) and lean and rich loading were predicted close to plant data. However, prediction of reboiler duty was found to be around 20% higher than that of the plant. This is due to the low estimation of lean/rich heat exchanger overall heat transfer coefficient. It can be inferenced here that steady state value of this need to be increased to reach the desired inlet temperature to the desorber.

Moreover, as discussed in previous section, the performance of the equilibrium-based absorber model of Tiller plant was found to be operating close to equilibrium with very high liquid to gas interaction as evident from the temperature profile obtained. Efforts also needs to be made to identify the reason behind high stage efficiency.

Table 4. Comparative results of simulation of Tiller pilot plant (SS: steady state simulation, Dyna: dynamic simulation).

Parameter	Pilot plant data [2]	Using rate based model at SS	Using equilibrium based model at Dyna (y=0.8)
Lean loading (mol CO_2/mol MEA)	0.14	0.15	0.15
CO_2 captured from gas (kg/hr)	15.30	14.75	14.85
Rich loading (mol CO_2/mol MEA)	0.48	0.49	0.47
CO_2 desorbed (kg/hr)	14.80	14.01	13.98
Reboiler duty (kW)	18.5	18.5	19.2

These comparison with steady state plant operational data validates the performance of the equilibrium-based models with stage efficiencies lower than 1. It can be inferenced here that this method is capable of predicting accurate plant performance within a range below 3-6%. Moreover, the simulation time and computational complexity were found similar to that of using simple equilibrium-based models. This serves the purpose of choosing such models to be used with dynamic models of power plants. However, for more accurate and detailed study on equipment performance and characterization such as of absorbers still require to use rate-based and detailed models of their. Further, validation of this method using transient plant data can confirm the application for analyzing transient operation of such plant. In the next section, simulation of one such transient operation of Gløshaugen plant is presented.

4.2. Using transient operational data

Dynamic simulation of the plant model of Gløshaugen pilot plant during transient operation as step changes in solvent flow rate was performed with determined stage efficiencies in the models. Simulation results for absorption and desorption rate, and rich amine flow rate and loading were compared with operational data from pilot plants.

The plant was subjected to a step change in lean amine flow rate of 21.8%. This changed lean amine flow rate from 172.3 kg/hr to 209.9 kg/hr. Upon reaching a new steady state condition, the rich loading reduced as well the desorbed CO_2. This led to reduction of lean loading as well the amount of CO_2 absorption. As the plant utilizes the captured and desorbed CO_2 and recirculates them in the flue gas, eventually the mole fraction of CO_2 reduced during the operation. Therefore, this operational data allows operating the plant under different operating region with various CO_2 content in the flue gas. Simulation of this unique operation would certainly validate the capability of our model for predicting transient response of PCC plants.

Simulation of the dynamic model for case 2 was continued up till 60 minutes and then the step change in the lean amine flow rate was applied. As the flue gas was used as a boundary stream, it was required to supply the variation of CO_2 content directly. Though, for ease of simulation, exact variation of CO_2 content was not supplied, instead, a step change with the same value as found in plant data was applied at the same time the amine flow rate was changed.

Results of simulation were compared with that of the pilot plant and presented in Figure 9 and 10. It can be observed from the figures that an average deviation in simulation results were lower than 3% for all of the parameters noted. Also, the trends in changing the parameters were similar to that of the plant data. As in the cycle the CO_2 content reduced, the amount of absorbed and desorbed CO_2 also reduced as evident from the Figure 10. This scenario was properly reproduced by the simulation. However, sudden change in captured CO_2 can be observed in Figure 10.a. This is due to the application of step change in the CO_2 content in flue gas. Subjecting this change similar to that of the plant would have increased the delay in such change, which in this case is not reflected. In addition, the delay in settling rich loading (Figure 9.a) to a new steady state was observed to be high compared to the plant data. It is due to high estimation of the sump volume and dimensions. This led to higher dead time in the cycle and as the rich loading value was recorded after the sump, it differ from that of the plant data. Nevertheless, variation of rich flow rate (Figure 9.b) found to be showing similar trend as well as reached the new steady condition same time as the plant. The reboiler duty was also found similar as the plant with little variation from the earlier steady state condition was observed.

When the computational times for simple equilibrium-based models and equilibrium-based models with stage efficiencies were compared, no significant change was observed. Therefore, as an alternative to rate-based modelling for absorbers and strippers, equilibrium-based models with reduced stage efficiencies can reliably be used for dynamic process simulation. This can provide acceptable accuracy with much lower modelling efforts and computational time compared to rate-based models.

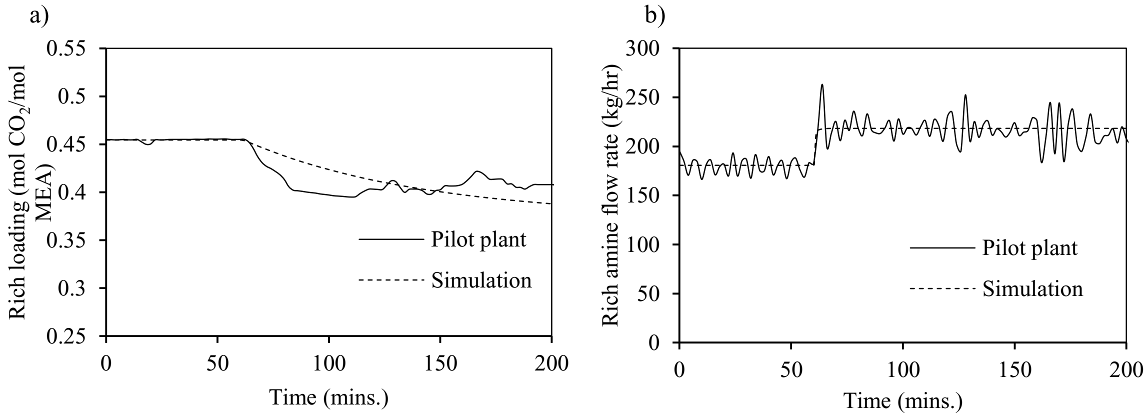

Fig. 9. a) Variation of rich loading and b) variation of rich amine flow rate: after applying step change in lean amine flow rate in the Gløshaugen pilot plant.

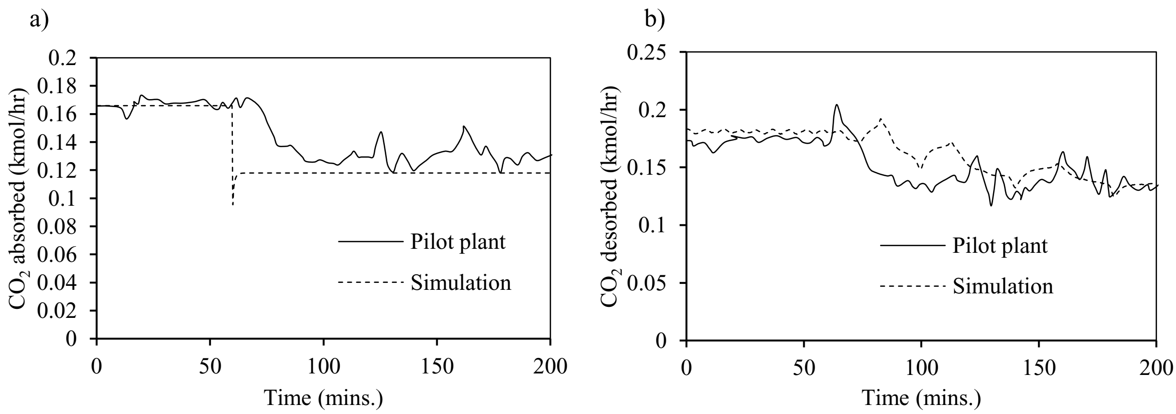

Fig. 10. a) Variation of absorbed and b) variation of desorbed CO_2 flow rate: after applying step change in lean amine flow rate in the Gløshaugen pilot plant.

5. Conclusions

Dynamic simulation of PCC processes are of interest to study the operability and control of such plants. However, it is a question of concern that how much accuracy for PCC plant models require when they are used along with the power plant models for analysing certain transients in the power plants as well as used in advanced model based controllers. As earlier researchers described rate-based models to be accurate enough to predict the performance of columns in the PCC plants, however, it takes considerably high time to simulate such processes. Though equilibrium-based models predicts inaccurate column temperature profiles, high capture rate at absorber, high reboiler duty etc., these type of column models take much low time to simulate such processes. In this paper, the use of reduced stage efficiency with equilibrium models was analysed for improved accuracy of such equilibrium models and for the purpose of validation, two different pilot plant data were used. It was found that prediction of end point conditions of such processes using such models is very much similar to that predicted by rate-based models and within a range of deviation of 3-6% as compared to the pilot plant data. Similar absorber temperature profiles also were found for the pilot plant with higher stage volume. Therefore, where we require to have faster models, equilibrium-based models with known deviation from actual results are an alternative to rate-based models instead of using reduced order rate-based models.

Moreover, following inferences may be drawn from this study.

1. Reduced stage efficiency in equilibrium models of columns can be used for integrating with power plant models for analysing the flexibility of such integrated plants.
2. Values of reduced stage efficiencies change with the packing type and absorber size. However, they do not change with the changing operating conditions.
3. Accurate steady state models can be used for determination of reduced stage efficiencies. It eliminates the requirement of existing plant or pilot plants for new PCC plant analysis.
4. Estimation of heat transfer coefficients requires verification before it can be used in dynamic simulation in order to obtain accurate performance of the desorbers.
5. In order to optimize the performance of a PCC model, the deviation in capture rate can be used as the parameter in a single parameter optimization.

Further study needs to be performed in order to identify how the stage efficiency changes with changing packing material and absorber sizes. It is also required to investigate the reason of high liquid and gas interaction with increased stage volume and the trade-off between the absorber pressure drop and stage volume. Furthermore, PCC models with reduced stage efficiencies need to be integrated with dynamic models of power plant and require to evaluate their performance in terms of accuracy of the simulation results, overall computational complexities and simulation time.

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