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Dynamic simulation and analysis of a pilot-scale CO₂ post-combustion capture unit using piperazine and MEA

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Abstract: Post-combustion capture is a promising technology for developing CO₂ neutral power plants. However, to make it economically and technically feasible, capture plants must follow the fast and large load changes of the power plants without decreasing the overall performance of the plant. Dynamic modeling and simulation is therefore needed to evaluate the performance of this plant under critical operation.

In this work, we evaluate the transient response of an absorber and a desorber for step changes of key process parameters, e.g. flue gas flow and composition, lean and rich CO₂ loading, etc. We show the results for the baseline 30 wt% MEA and the low energy piperazine (PZ) solutions. This analysis reveals that the absorber reaches steady-state faster using MEA compared to PZ. This is related to the shift of the mass transfer zone due to changes in temperature. The transient operation in the regeneration unit is somewhat similar while using both solvents: an initial fast decrease of the lean loading is followed by a slow transient period as the system approaches steady-state conditions. We show the presence of inverse response in the stripper column when the rich loading decreases or the feed's temperature reduces using PZ solvent. Thus, we demonstrate that the dynamics of the MEA system cannot be extrapolated to other solvents.

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Keywords: Modelling and System Identification; Process Optimization and Plant wide Control; CO₂ post-combustion capture;

1. INTRODUCTION

Although great efforts have been placed for the rapid growth and development of the renewable energy market, thermal power plants still represent the world's main energy supply and, especially in developing countries, they will dominate the market in the coming decades (Hammond et al., 2011). Fossil-fuelled power plants produce almost one third of the global CO₂ emissions (Pachauri et al., 2008); thus, CO₂ capture would greatly reduce the impact of power plants on the climate.

Post-combustion capture is one of the leading technologies in CO₂ capture. It is a mature concept, ready to be implemented on a large scale (Bui et al. 2014). However, plant-wide dynamic studies are needed to further demonstrate the feasibility and flexibility of thermal power plants with integrated CO₂ capture. Power plants need to handle fluctuations resulting from various sources, such as peak in energy demands, change in green energy production, raw material heterogeneity, malfunctioning of equipment, etc. As a consequence, CO₂ capture units need to accommodate large load changes or eventual shutdowns to become attractive.

Recent efforts in the CO₂ capture field have focused on dynamic model development and flexibility evaluation of different operational scenarios (Sahraei and Ricardez-Sandoval 2014, Nittaya et al. 2014). In the context of dynamic modelling, some of the recent contributors which are particularly noteworthy are (Harun et al. 2012, Lawal et al. 2010, Mac Dowell, 2013). They presented dynamic models for the absorber and for the desorber and investigated the effect of various changes on the performance of the capture plant. These studies represent a first insight into the dynamics of post-combustion capture plants.

The majority of these studies have used monoethanolamine (MEA) as solvent. To our knowledge, only three studies present dynamic models using other solvents. Gaspar and Cormos (2012) presented an absorber model for MEA, diethanolamine (DEA), 2-amino-methylpropanol (AMP) and methyl-diethanolamine (MDEA) solvents. They demonstrate that kinetics play a key role in the dynamic behaviour of the capture process. Walters et al. (2013) presented a first-principle based dynamic model for the alternative two-stage flash stripper configuration with piperazine (PZ) assuming an

equilibrium stage process. Gaspar et al. (2015a) implement a dynamic rate-based model using MEA and PZ.

The purpose of this paper is to explore the dynamic behaviour of an absorber and a desorber using 30 wt% PZ and to compare the dynamics to the 30 wt% MEA for step changes in key operational parameters, e.g. flue gas flow rate, flue gas composition, lean and rich CO₂ loading and rich feed temperature. The dynamic CAPCO₂ (dCAPCO₂) in-house DTU model was employed to describe the behaviour of CO₂ absorption and desorption (Gaspar et al., 2015a, b). The present analysis provides a first insight into the dynamics of the columns using the innovative PZ solvent. Furthermore, we highlight the similarities and differences between PZ and MEA and we indicate some of the operability/control aspects which need to be considered when using different solvents.

2. THE DYNAMIC CO₂ CAPTURE MODEL

In the present work, a mechanistic first-principle based dynamic mathematical model for CO₂ absorption and desorption (dCAPCO₂) is used for the simulation of the absorber and the desorber (Gaspar et al. 2015a). The partial differential equations (PDE) describing the spatial and temporal variation of state variables are coupled with algebraic equations for mass and hydraulic properties (Rocha et al. 1993, 1996), mass and heat transfer fluxes, and the extended UNIQUAC thermodynamic model (Thomsen & Rasmussen, 1999). The PDEs are discretized in the axial domain using the finite differences method. Therefore, the model becomes a system of ordinary differential and algebraic equations (DAE), with time as the independent variable. This set of DAE is integrated using the ODE15s Matlab solver. This model has been validated against pilot experimental data using MEA (Gaspar et al. 2015b). Validation of this model using PZ solvent is discussed below.

2.1 Validation of the Piperazine CO₂ Capture Model

We compare the model predictions to experimental measurements for CO₂ absorption and desorption against pilot plant data carried out at the J. J. Pickle Research Center, north of Austin, TX, USA. We include campaigns “Fall 2008” and “Fall 2010”. A detailed description of the experimental setup and operation conditions is presented by Van Wagener (2011).

Fig. 1 and 2 show the agreement between the model and pilot results for absorption and desorption. Fig. 1 illustrates that the calculated and measured CO₂ absorption percentages are in reasonable agreement. The deviations between the model predictions and the experiments are less than 10%. Plaza (2011) outlines that the accuracy of the absorber titrations are within $\pm 10\%$ and the liquid side removal matches the gas side results within $\pm 15\%$. Thus, the model predictions are within the accuracy of the measurements.

There is only one point visibly outside of the $\pm 10\%$ range which is most probably an outlier (see Fig. 1). The flue gas inlet temperature for this point was -5°C , which represents the lower limit of the experimental temperature range.

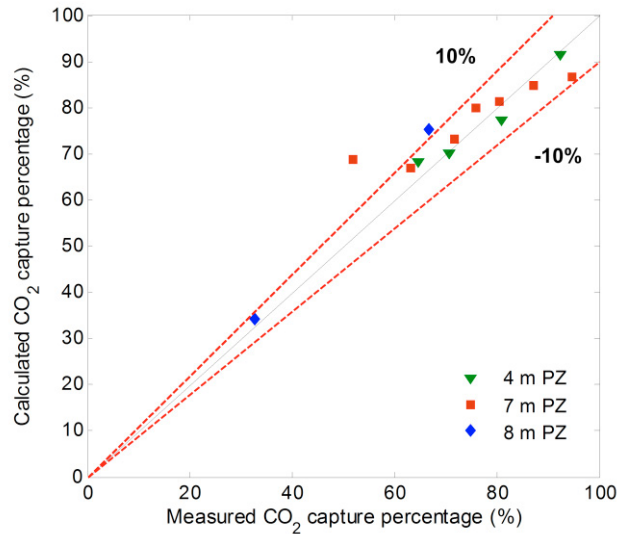


Fig. 1. Calculated versus measured CO₂ capture percentage – campaign “Fall 2008”

This might lead to low water content and it could be the reason for over-prediction of the model.

Fig. 2 presents the calculated and measured lean CO₂ loading for the campaign “Fall 2010”. The agreement between the model and pilot results is good. The predictions are in the $\pm 10\%$ range and the error in the prediction is not systematic.

Fosbøl et al. (2014) investigated the expected uncertainty of some calculated simulation properties. They concluded that 5-10% deviations are expected when comparing the model predictions to experimental data. Some properties, such as rich loading, outlet temperatures, CO₂ product purity are more reliable than other properties such as CO₂ capture percentage, stripper top CO₂ flow, reboiler temperature, etc. Therefore, it can be concluded that the predictions are in good agreement with the pilot measurements.

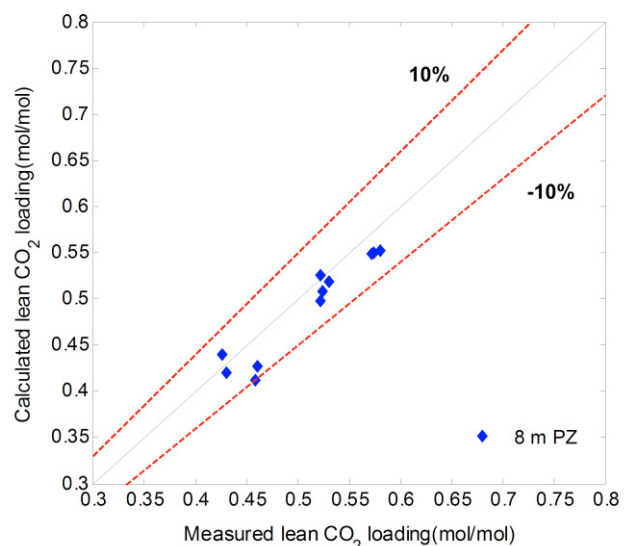


Fig. 2. Calculated versus measured CO₂ lean loading – campaign “Fall 2010”

3. DYNAMIC SIMULATION AND ANALYSIS

One of the major costs associated with the operation of a CO₂ post-combustion capture plant is the circulation and regeneration of the solvent. They are mainly influenced by the performance of the absorber and the desorber units. Accordingly, we focus first on the transient response of the absorber for step changes in the flue gas flow rate, flue gas composition and lean CO₂ loading. Afterwards, we analyse the dynamics of the stripper for variation of the CO₂ loading and temperature of the feed stream. This study represents the first step towards the development of suitable control strategies for the piperazine-based CO₂ capture plant.

3.1 Process Design

The base case operating conditions correspond to a 1 t/hr CO₂ capacity post-combustion capture plant using 30 wt% MEA (7 molal) and 30 wt% PZ (5 molal) solutions, respectively. The loading of the lean solution entering the absorber is approximately 0.20 mol CO₂/mol alkalinity at 40°C. The flue gas coming from a coal-fired power plant contains 12.4 mol% of CO₂ and it is saturated with water before entering the absorber. We assume that the SO_x and NO_x content of the flue gas is removed upstream of the capture process. The plant is designed for 90% CO₂ removal. Table 1 summarizes the main design specifications and process parameters for the post-combustion CO₂ capture plant (Faber et al., 2011).

Table 1. Design specifications for the absorber and the stripper

Flue gas flow rate (mol/s)	61.5
Flue gas temperature (°C)	40
Flue gas pressure (kPa)	101.32
Flue gas CO ₂ composition (mol%)	12.4
Flue gas H ₂ O composition (mol%)	10.9
Lean inlet temperature (°C)	40
PZ/MEA lean loading (mol/mol alk.)	0.18/0.2
Amine concentration (wt%)	30
L/G ratio for PZ/MEA (mol/mol)	3.5/3
CO ₂ recovery (%)	90
Column diameter (m)	1.1
Absorber/Desorber height (m)	17/10
Reboiler operating pressure (kPa)	185

3.2 Absorber Simulation

This section shows the dynamic behaviour of the absorber for three scenarios: ±10% step change in the flue gas CO₂ concentration (case 1), ±10% step change in the lean CO₂ loading (case 2) and ±10% step change in the flue gas flow rate (case 3). These steps are applied to the base case after 10 min of steady-state operation. Here, we show the results for both solvents: PZ and MEA.

In practice, case 1 resembles operational conditions when the output of the power plant changes due to the heterogeneity of the fuel. This case is common, especially for biomass co-fired power plants. Case 2 resembles a scenario when a

disturbance occurs in the operation of the stripper, e.g. steam supply shortage. Case 3 corresponds to part load operation of the power plant and represents one of the most common scenarios observed during flexible operation. Case 1 and case 2 result in changes of the CO₂ concentration gradient between the gas phase and the liquid phase. This gradient represents the driving force for absorption. Case 3 results in varying contact time inside the column, which changes the L/G ratio between the gas and the liquid phases.

Fig. 3 shows the dynamic performance of the absorber for each case study using MEA and PZ. This figure illustrates how an increase of the flue gas CO₂ content, lean CO₂ loading, or the flue gas flow rate results in a reduction of the CO₂ capture efficiency and vice-versa (case 1 to 3). Furthermore, it highlights that the effect of a step change is greater using PZ compared to MEA. A 10% decrease of the shown variables results in a CO₂ capture percentage of approximately 92% and 96% for MEA and PZ, respectively. For a 10% increase, the CO₂ capture percentage reduces to 86% and 83% for MEA and PZ, respectively.

Figure 3 also indicates that, for all the cases, the MEA system reaches steady-state faster than PZ. The CO₂ capture percentage stabilizes in about 10-15 minutes using MEA and using PZ stabilizes in roughly 40 min for a 10% step increase and in about 1 hour for a -10% step change. This is contrary to expected since PZ has a faster kinetics than MEA (Dugas, Rochelle 2011).

It was shown by Gaspar and Cormos (2012) that solvents with fast kinetics respond faster than those with slower kinetics. To understand this behaviour, the dynamics of the absorber for -10% step change in the flue gas CO₂ content is discussed in detail (Fig. 3A). Detailed analysis of case 2 and case 3 are not shown here for brevity; however, the dynamics of the system is similar to case 1. We chose the -10% step scenario since the difference between PZ and MEA is the most visible compared to the other cases.

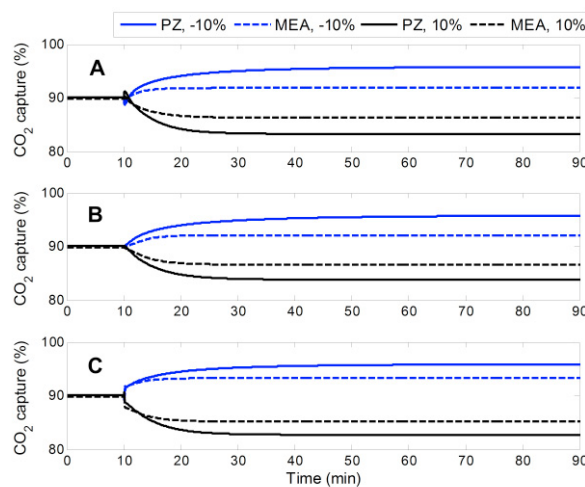


Fig. 3. CO₂ capture percentage versus time using MEA and PZ for (A) case 1 – CO₂ composition, (B) case 2 – Lean loading and (C) case 3 – flue gas flow.

Case 1: Decrease of the flue gas CO₂ composition

The case study presented here is equivalent to reducing the amount of CO₂ available for capture and it consists in lowering the driving force for absorption. The focus is on defining the differences between PZ and MEA and describing the dynamic coupling between temperature, gas phase composition, and liquid phase composition. These results are shown in Fig. 4 to 6.

Fig. 4 presents how the gas phase CO₂ concentration decreases as function of time at several locations in the column for both MEA and PZ solvents. Fig. 3A also shows how the CO₂ capture percentage increases. This is expected, since the free-amine to CO₂ gas ratio increases when the CO₂ composition is reduced. Furthermore, Fig. 4 illustrates that the MEA system responds faster since it reaches steady-state within 10 minutes. Contrary to MEA, PZ responds much slower. The settling time for the outlet top CO₂ concentration ($H_c=17\text{ m}$) is roughly 30 min; however, the CO₂ concentration in the middle section of the absorber reaches steady state much slower, in approximately 2 hours. Nevertheless, both solvents present an initial fast response followed by a slow transient period as the system approaches the new steady-state.

Gáspár and Cormoş (2011) and Mac Dowell et al. (2013) showed that, for the MEA, the dynamics of mass transfer is tightly coupled to temperature changes and vice-versa. This is expected since additional CO₂ absorption by MEA produces more heat. On the other hand, higher temperature results in greater evaporation rate and it is favourable from a kinetic point of view. This coupling is obvious by comparing Fig. 4 to 6 as discussed below.

Fig. 5 presents the CO₂ absorption rate as function of the column's height. Fig. 5A on MEA outlines the presence of a small bulge in the absorption rate, but the efficiency of the column is well-balanced, between 0.1 and 0.2 mol CO₂/s alongside the height of the column. Contrary to MEA, there is a visible peak in Fig. 5B on PZ. This peak results in: (1) a more efficient section (4-7 m long, with an average absorption rate of 0.4 mol CO₂/s) where most of the CO₂ absorption takes place and (2) two less efficient sections above and below the mass transfer bulge (absorption rate below 0.15 mol CO₂/s). Fig. 5B shows that this bulge moves downwards as a consequence of the 10% step decrease in the CO₂ content.

To provide further insight on the dynamics of mass transfer, we present the temperature profile inside the column at selected time snapshots in Fig. 6. This figure reveals how the temperature profile is almost constant for MEA and it stabilizes in less than 10 min, similar to the gas phase composition in Fig. 4. The PZ temperature changes more slowly than the corresponding MEA temperature profiles. Fig. 6B shows that the PZ temperature bulge gradually expands over the middle section of the absorber.

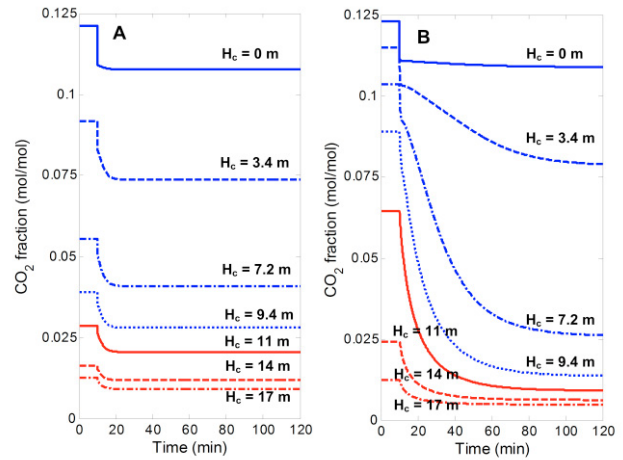


Fig. 4. Gas phase CO₂ composition versus time using (A) MEA and (B) PZ.

By inspecting Fig. 5B and 6B on PZ, one can observe that the shift of the temperature bulge produces a change in the CO₂ absorption rate. Before the step change ($t=10\text{ min}$), the location of the peak is at 11.5 m while the temperature bulge is located around 14 m. As the system evolves, the mass transfer peak gradually moves to $H_c=5\text{ m}$, while the bulge expands over 7 m of height.

The results from this analysis show that the slow change of the temperature is synchronized with the shift of the mass transfer peak from the top to the bottom of the column. The absorption rate strongly depends on the temperature and it may result in a mass transfer pinch, as reported for steady-state conditions by Sachde, Rochelle (2014).

This analysis reveals that piperazine responds slower to disturbances than MEA and the inlet parameters have a significant effect on the PZ process. Accordingly, feedback controllers with high gains and short time-integrals may be required in the case of PZ to maintain the dynamic operation of the absorber column within reasonable short closed-loop settling times in the presence of these disturbances.

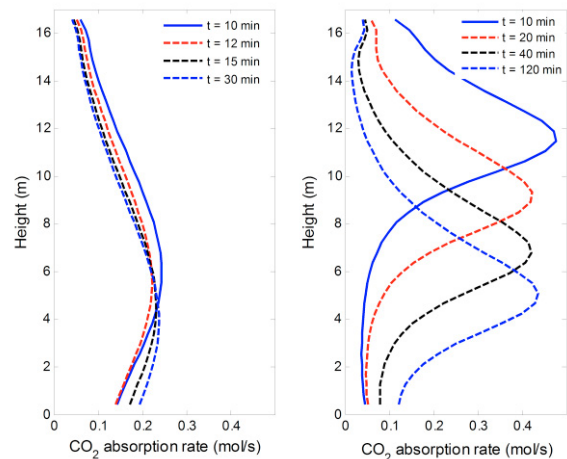


Fig. 5. CO₂ absorption rate versus height using (A) MEA and (B) PZ.

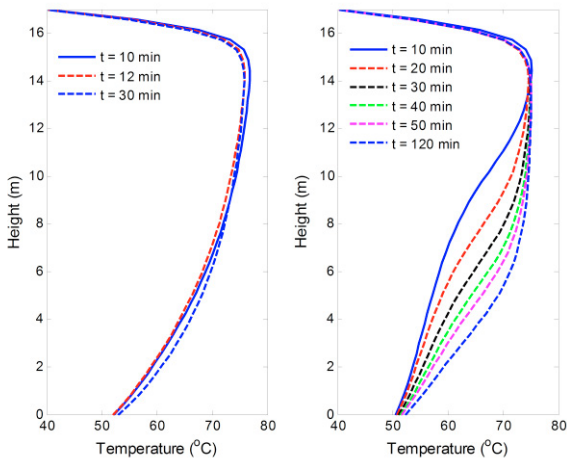


Fig. 6. Temperature versus absorber's height using (A) MEA and (B) PZ.

3.3 Desorber Simulation

We analyse the transient behaviour of the stripper using MEA and PZ for two scenarios: ± 0.02 mol/mol alk. step change in the CO₂ loading of the rich feed (case 4) and $\pm 1^\circ\text{C}$ step change in the temperature of the rich feed (case 5). The reboiler heat duty is kept constant during this analysis and the step is applied after 10 min of steady-state operation. Case 4 corresponds to small disturbances in the operation of the absorber. It may result from changes in the flue gas stream, as demonstrated above in cases 1 to 3. Case 5 resembles a disturbance in the operation of the absorber or the lean-rich cross-heat exchanger.

Fig. 7 shows the dynamic response of the stripper for cases 4 and 5. It outlines that a step change response of the stripper is to some extent similar between MEA and PZ. Generally, the transient behaviour is a sudden decrease of the lean loading followed by an increase towards steady state. However, some differences between the cases are noteworthy: (1) Steady-state is reached within 10–15 min when rich loading decreases respectively rich temperature reduces, except for the $+1^\circ\text{C}$ step change using PZ, then steady-state is reached within 30 min (Fig. 7B). (2) A greater rich loading respectively lower rich temperature reduces the performance of the stripper using PZ and improves the stripping efficiency using MEA. The settling time is around 30-40 min.

The simulation results (not shown here) demonstrate that the same behaviour is observed for slightly larger step changes of ± 0.03 in loading and $\pm 4^\circ\text{C}$ in the temperature of the rich feed.

Fig. 7 demonstrates the phenomenon of inverse response of the stripper for the PZ solvent when: (case 4a) rich loading increases (Fig. 7A, blue line) and (case 5b) feed temperature decreases (Fig. 7B, red line). Inverse response of a unit arises from competing dynamic effects that operate on two different time scales. Systems that present this behaviour are challenging to control.

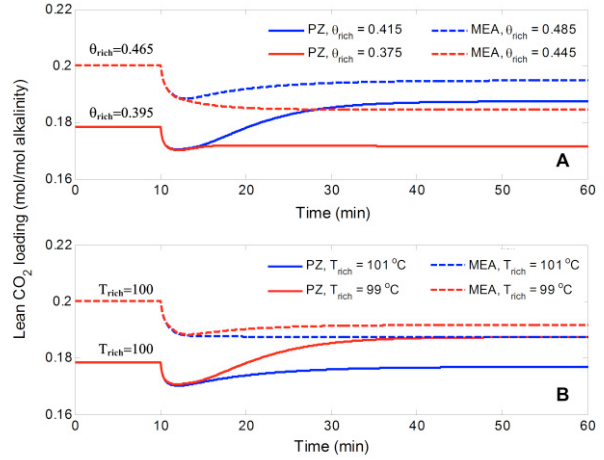


Fig. 7. Lean CO₂ loading for (A) case 4 – rich loading and (B) case 5 – feed temperature

We exemplify in Fig. 8 the two competing effects resulting in inverse response of the stripper for cases 4a and 5b. Fig 8A shows that the solvent flow rate to the reboiler increases instantaneously from 188.3 mol/s to 189.3 mol/s for cases 4a and 5b, respectively. As a consequence of the greater solvent hold-up, the reboiler outlet vapour flow increases 0.3 mol/s in 1 min (see Fig 8B). The increased boiling produces a leaner reboiler stream, which corresponds to more efficient stripping (Fig. 7).

However, the initial efficiency improvement is followed by a slow transient decrease. Fig. 8 shows that the solvent flow to the reboiler, and consequently the reboiler boil-up, reduces below their initial values within 8-15 min. Lower boil-up rate produces lower CO₂ desorption rate.

It can be concluded that the transient behaviour of the MEA system cannot be extrapolated to other solvents. Dynamic models are necessary to fully understand the transient behaviour of a capture plant and to design robust control structures.

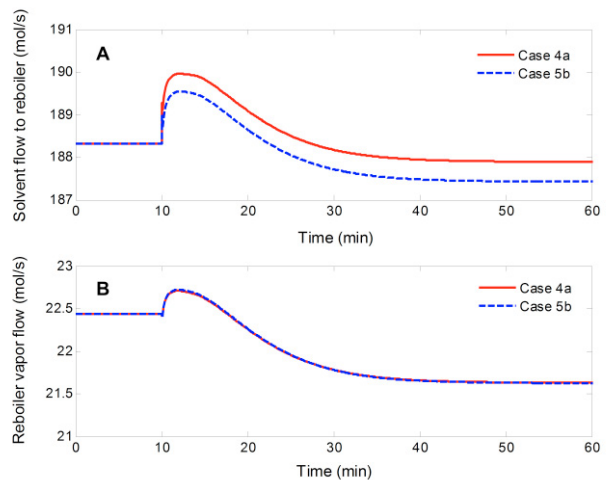


Fig. 8. (A) Stripper outlet solvent flow and (B) Reboiler boil-up for cases 4a and 5b using PZ.

4. CONCLUSIONS

This study shows a dynamic analysis of a 1 t/h CO₂ capacity absorber and desorber using PZ and MEA for changes in the flue gas composition and temperature, the lean solvent flow rate respectively the temperature and CO₂ loading of the stripper's feed. We have demonstrated that step changes in the flue gas and the lean flow rate have a significant impact on the absorber. The settling time is approximately 2–3 times slower in case of PZ compared to MEA. This behavior is related to the coupling between temperature and mass transfer rate. The analysis of the desorber revealed a strong influence of the rich loading and feed's temperature on the lean CO₂ loading. We have shown that a step change in the composition or the temperature of the stripper's feed produces a sudden decrease of the lean loading followed by a slower transient period when approaching steady-state, for both solvents (MEA and PZ). Thus, the desorber evolves on a fast and a slow time-scale. We have outlined the presence of inverse response for increased rich loading or decreased feed temperature. The observed initial decrease in the outlet lean loading is later overcome by a decrease in the vapor boil-up. Current work in this research is focused on developing a plant-wide control study using industrially-relevant scenarios, e.g. typical changes in the load, malfunctioning of equipment, limited heat supply, and to evaluate the effect of design variables on process dynamics, e.g. size of storage tanks and sumps, dimensions of the absorption and desorption towers and heat exchangers.

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