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Design, economics and parameter uncertainty in dynamic operation of post-combustion CO_2 capture using piperazine (PZ) and MEA

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Abstract

Post-combustion capture is a promising solution to mitigate the anthropogenic CO_2 emission rate and reduce global warming. However, to make it economically attractive, the techno-economic performance of this process needs to be improved. This includes steady-state but also dynamic operation of the plant. Flexibility is particularly crucial from an economic and operational point of view since plants must balance the power production and the electricity demand on a daily basis.
This work shows the impact of design decisions and uncertainties on the dynamic operation and economics of a CO_2 capture plant using piperazine (PZ), compared to the benchmark MEA solvent. This is exemplified through dynamic model calculations. The results show that the capacity of the buffer tank is a key parameter for the flexibility of the plant. A small tank corresponds to lower capital cost but it leads to increased operation cost and also to flexibility/controllability issues. Both, the PZ and MEA plants present inverse response for small tanks. These plants are challenging to control.

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Keywords: Dynamic design; Economics; Flexibility; Uncertainty; Piperazine;

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1. Introduction

It is becoming widely recognized that mitigation of anthropogenic CO_2 is essential to reduce global warming [1]. One of the potential short and mid-term solutions for CO_2 emission reduction is solvent based CO_2 capture. Despite the efforts and recent advances in the development of renewable energies, the energy infrastructure is not ready to replace the fossil-fuel fired power plants with renewables. Thermal power plants represent the main energy supply and they are expected to dominate the market in the coming decades, especially in developing countries [2]. As a result, the growing focus on CO_2 emissions mitigation requires integration of fossil-fuel fired power plant with CO_2 capture units. Post-combustion CO_2 capture (PCC) has emerged as one of the main alternatives for CO_2 capture and it is moving towards industrial deployment. PCC is a mature technology that can be implemented in existing power plants and it is also suitable for other sectors, e.g. the steel industry, cement production, petroleum refining, and the biochemical industry.

Most of the research on CO_2 capture has been focused on steady-state optimization, design and techno-economic evaluation, generally applied for the baseline mono-ethanol-amine (MEA) [3]. Dynamics of the plant is considered only after the design specifications and operating variables have been determined from the steady-state analysis, e.g. packing height, solvent flow rate, heat demand. However, a PCC plant must be able to handle fluctuations resulting from various sources, such as peak in energy demands, raw material heterogeneity, malfunctioning of equipment, increasing share of renewable energy sources, etc.

Flexibility is particularly crucial from an economic and operational point of view. Plants must balance the power production and the electricity demand on a daily basis [4]. Therefore, the sequential steady-state design and controllability analysis approach may result in a dynamically inoperable capture plant since the design of the plant imposes a limitation on the process dynamics and therefore on the flexibility of the plant. To develop a flexible and economically efficient post-combustion unit, these should be designed considering the transient evolution of the plant and not only the steady-state behavior [5,6]. Furthermore, there is a gap of knowledge in the dynamics and controllability of novel-low energy solvents since most of the research have focused on the benchmark MEA capture process. An example of a promising solvent is the 5 molal piperazine solution which offers higher CO_2 capacity and an energy improvement of approximately 20% compared to MEA [7]. However, rate-based models using novel solvents are less reliable due to insufficient kinetic and physico-chemical data; thus, uncertainties must be accounted for when designing and evaluating the controllability of a CO_2 capture plant.

The aim of this work is to investigate the importance of design variables such as the capacity of buffer tanks, sumps, lean-rich heat exchanger, etc. on the transient operation of a CO_2 capture plant using the low energy piperazine (PZ) and compared its performance against the benchmark MEA solvent. Here, we focus on how the capacity of the buffer tank influences the dynamics of a pilot-scale plant since the sizing of this unit is generally omitted in controllability studies. In addition, we discuss how the capacity of buffer tank influences the economic performance of the post-combustion capture unit. This is illustrated through the capture percentage and the specific reboiler duty. These parameters are related to the dynamic variability and operational cost of the capture plant.

The results of this work were developed using the dynamic CAPCO2 (dCAPCO2) in-house DTU model for CO_2 absorption and desorption. The complete capture process has been implemented in Matlab and this model has been validated against experimental data [8,9]. However, models are not perfect and a key aspect in assessing the dynamic techno-economic performance of a plant is quantification of expected uncertainties, e.g. kinetics, mass transfer and hydraulic models [10]. Here we exemplify the effect of uncertainties related to the kinetic model in the dynamics of the capture process.

2. Modeling of CO_2 Post-Combustion Capture

2.1. Dynamic model description

This section briefly discusses the dynamic model of the pilot-scale post-combustion capture plant. This model of the plant consists of an absorber, a desorber and auxiliary units, i.e. sump, the buffer tank, the reboiler and the heat exchanger, as shown in Fig. 1. The absorber and the desorber are represented by the DTU dCAPCO2 dynamic rate-based model. This model takes into account the accumulation of mass and energy in both the gas and the liquid

phase and it uses the general model (GM) enhancement factor to account for the simultaneous reactions and mass transfer phenomena [11,12]. The kinetic model includes two parallel reactions to describe the CO_2-PZ system and one single reaction to characterize the CO_2-MEA system. It is essential to include both of the reactions for the PZ solvent since the contribution of the bicarbamate forming reaction is greater than 30% at high CO_2 loading [11]. Furthermore, the dynamic model uses the extended UNIQUAC thermodynamic model to calculate vapor-liquid equilibrium, thermal properties and speciation in the electrolyte systems [13]. The model of the complete post-combustion plant is implemented in Matlab in combination with FORTRAN subroutines for mass transfer, hydraulic and thermodynamic model calculations. The material and energy conservation equations were discretized in the axial domain using the finite differences method (FDM). FDM represents the spatial derivatives in discrete grid points. Therefore, the partial differential equations system reduces to 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 with the boundary conditions at the top for the liquid phase and at the bottom for the gas phase. This approach provides a simple way to solve the model. However, realistic initial conditions have to be provided to obtain convergence. To assure fast convergence and robustness, the dynamic model is initialized using steady-state values, determined by the CAPCO2 steady-state model [14–16]. The models of the absorber sump and of the reboiler are simplified in this study. A dynamic continuous stirred-tank model is used for simulation of the absorber sump [17]. The reboiler and heat exchanger is modelled as a flash tank with instantaneous heat exchange between the heating medium and solvent assuming perfect level control, similar to [18].

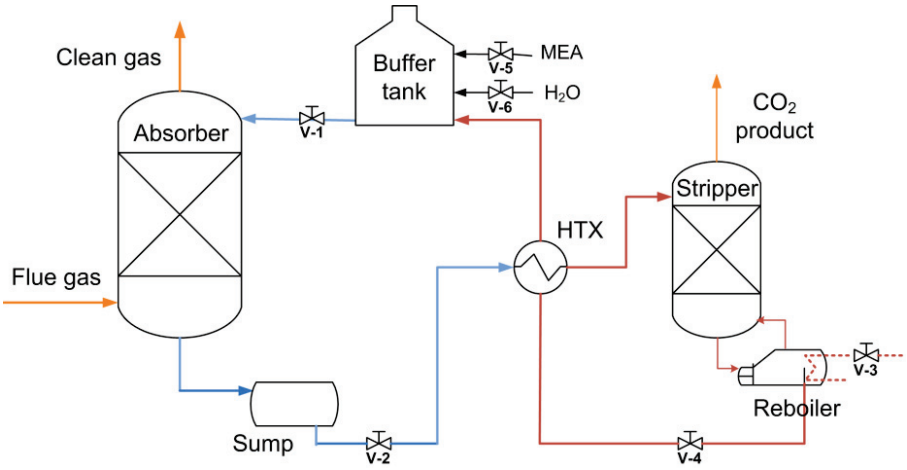

Fig. 1. Post-combustion CO_2 capture process flowsheet.

The developed CO_2 capture model was compared to dynamic experimental data using MEA and it was compared to steady-state pilot plant data using PZ [8,9,19,20]. Additionally, 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 CO_2 loading, etc. was compared using amines PZ and MEA [21]. These studied showed that the applied model predicts well the transient evolution of the capture plant for operationally relevant scenarios, e.g. changes in the load, lean solvent and steam flow rate. It captures the fast responses in the columns as well as the slow transient evolution with time delays. Therefore, this model and its numerical implementation enable transient simulation of a pilot-scale post-combustion capture plant and it is adequate for dynamic behavior analysis and control strategies development.

2.2. Design specifications and operating conditions

The design base of the plant is the process configuration with heat integration between the lean and the rich solution. This configuration resembles the pilot plant reported in a previous study [22]. The nominal operating parameters and the design specifications for the PZ respectively MEA plant originate from [21] and they are summarized in Table 1. The simulated capture plant removes 1 t/h CO_2 from the exhaust of a coal fired power plant. The flue gas contains 12.4 mol% CO_2 and it is saturated with water at the absorber inlet temperature of 40°C. Moreover, we assume that the flue gas is treated for control of post-combustion products and therefore it consists of inert gases (mixture of N_2 and O_2), CO_2 and H_2O. It is assumed that the inert gas has the same characteristics as air. The amine and CO_2 composition of the lean solvent was determined to capture 90% of the inlet CO_2 [23]. These values assure comparable liquid hold-ups between the PZ and the MEA plant and they corresponds to conditions near the optimal operating range.

Table 1. Main nominal operating parameters and design specifications for the CO_2 capture plant

Input parameters	Units	PZ	MEA
Flue gas flow rate	mol/s	61.5	61.5
Flue gas temperature	°C	40	40
Flue gas CO_2 composition	mol%	12.4	12.4
Lean solvent flow rate	mol/s	195	251
Lean inlet temperature	°C	40	40
PZ/MEA lean loading	mol/mol	0.34	0.19
Amine concentration	wt.%	30	30
Reboiler operating pressure	kPa	185	185
Heat exchanger temperature approach	°C	10	10
Column diameter	m	1.1	1.1
Absorber/Desorber packing height	m	17/10	17/10
Absorber/Desorber sump diameter	m	1.1	1.1

The absorber and the stripper are packed columns equipped with Mellapak 2X structured packing and IMTP50 dumped packing, respectively. The absorber sump and reboiler tank are designed to allow large solvent flow rate changes during flexible operation of the capture plant and the heat exchanger operates with a lean-rich temperature difference of 10°C.

3. Design, Economics and Parameter Uncertainty in Dynamic Operation

This section presents dynamic simulation results with focus on the effect of design variables on the transient response (flexibility) and economics of the PZ and MEA plants, respectively. First, this section shows how the capacity of the buffer tank, i.e. the nominal volume of the liquid in the “Buffer tank” in Fig. 1, influences the transient evolution of the CO_2 capture plant using PZ and MEA in presence of step changes in key operating parameters, such as the flue gas flow rate, the solvent flow rate and the steam flow rate (reboiler duty). Changes in these key parameters occur frequently as power stations must operate under a wide range of operating conditions to balance between energy production and demand. The focus of this analysis is on the CO_2 removal efficiency and the heat needed to regenerate the CO_2 rich solvent (specific reboiler duty). Furthermore, this section shows the effect of an uncertainty of ±10% in the kinetic model in the dynamic response of the capture process.

3.1. Design, flexibility and economics in dynamic operation

The first part of the dynamic analysis focuses on the transient response of the PZ respectively MEA plant with different buffer tank capacities for step changes in key parameters, i.e. flue gas flow rate, lean solvent flow rate and steam flow rate. This dynamic sensitivity analysis is performed in open-loop control. The capacity of the buffer tank corresponds to the nominal (initial) solvent volume (V) in the “Buffer tank”, Fig. 1.

In this analysis, all the parameters are kept at their nominal values as shown in Table 1 and only one parameter is step-changed at a time to investigate its effect on the transient response of the plant. The buffer tank capacity

(volume, V) in this work is set to 1, 3 and 12 m^3 for the PZ plant and 1.5, 4.5 and 18 m^3 for the MEA plant, respectively. The volume of the buffer tank for MEA is larger than for PZ as the MEA lean solvent flow rate is greater (251 mol/s) than the PZ solvent flow rate (195 mol/s), as shown in Table 1. The upper limit for the volume of tank, i.e. 12 m^3 respectively 18 m^3 allows for large solvent flow changes whereas the lower limit for the volume, i.e. 1 m^3 respectively 1.5 m^3, was selected to minimize the damping effect of the buffer tank.

This work also presents a rough estimate on the effect of the buffer tank capacity on the economics of the process. For example, Fig. 2 to Fig.4. show the CO_2 capture percentage and the specific reboiler duty for the scenarios considered in this analysis. A deviation in the CO_2 capture rate from the 90% set-point will result in additional cost due to the CO_2 tax, i.e. we assume that the CO_2 emission tax is zero when the capture rate is greater or equal than 90%, similar to the taxation of SO_x emission. The specific reboiler duty shows the heat used to regenerate 1 t of CO_2. It is a common metric used to quantify the energy performance of a capture plant. Note that a lower reboiler duty resembles a more efficient plant, as less steam is needed to remove the same amount of CO_2. Each of the transient scenarios considered in this work are described next.

Fig. 2 shows the response of the plant for ±10% step-change in the flue gas flow rate for different buffer tank capacities. This scenario corresponds to load changes of the power station which leads to variation of the flue gas flow rate. Fig. 2 outlines that the capacity of the buffer tank has no influence on how the capture plant responds to ±10% change in the flue gas flow rate. Fig. 2A1 and 2B1 show that higher flue gas flow results in lower CO_2 capture percentage and vice-versa, as expected. The change in the CO_2 capture percentage is roughly 5% for the PZ plant and it is approximately 3% for MEA, regardless of the capacity of the buffer tank. Additionally, Fig. 2A2 and 2B2 show that the response of the plant in the specific reboiler duty is inversely proportional with respect to variation of the flue gas flow, i.e. higher flue gas flow results in lower reboiler duty and vice-versa. However, the change in the reboiler duty is the same between different tank capacities for a given solvent. Note that the steam flow to the reboiler is constant during this scenario and the lower reboiler duty is due to the larger amount of CO_2 captured. Note in Fig. 2 that the MEA plant (Fig 2B) reaches the new steady-state faster than the PZ plant (Fig 2A). These findings are in accordance with the results reported in previous studies [19,21].

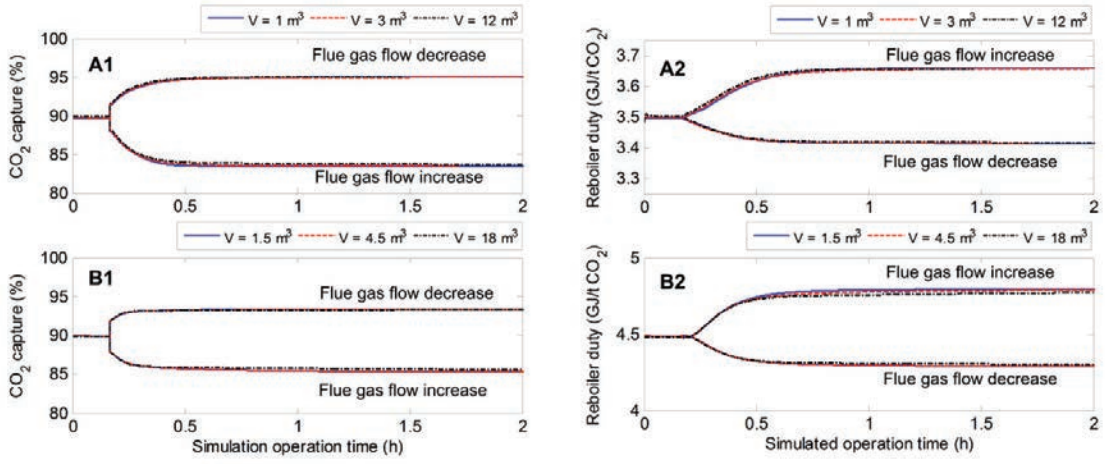

Fig. 2. Dynamic response of the plant for ±10% step change in the flue gas flow rate using: (A1) and (A2) PZ; (B1) and (B2) MEA.

Fig. 3 presents the dynamics of the PZ respectively MEA plant for 10% step-increase in the lean solvent flow rate for different buffer tank capacities. Note, the lean solvent flow rate is a key manipulated variable in the control of a post-combustion capture unit [5,21,24].

Fig. 3 illustrates that the capacity of the buffer tank has a strong effect on how the system responds with respect to the lean flow rate. Generally, a capture unit with a smaller buffer tank has a longer settling time, i.e. the time needed to reach the new steady state is longer when the volume of the tank is smaller for both solvents. Fig. 3A1 and

3A2 outlines that the PZ plant with a tank of 1 m^3 has a settling time of roughly 12 hours and the settling time is less than 4 hours for buffer tank volumes above 3 m^3. Regarding the behavior of the MEA plant, Fig. 3B1 and 3B2 show that its settling time is around 2 hours for a capacity of 1.5 m^3 and it is approximately 30 min for tank capacities above 4.5 m^3. This is important to consider when designing a capture plant: a small tank results in very long settling times and it may reduce the flexibility of the capture plant.

Furthermore, Fig. 3A1 and Fig. 3B1 outline the presence of inverse response in the CO_2 capture percentage for both, PZ and MEA plants for tank capacities of 1 m^3 and 1.5 m^3, respectively. A larger buffer tank eliminates the inverse response of the MEA plant (Fig. 3B1) but this behavior persists for the PZ plant, even for much larger tanks (Fig. 3A1). This is important to consider as systems with inverse response are challenging to control [25]. In addition, Fig. 3A2 and 3B2 show that a small buffer tank leads to large jump in the reboiler duty which corresponds to greater operating cost. Thus, a small tank corresponds to lower capital cost but it greatly increases the cost of operation (reboiler duty) and reduces the flexibility of the plant. The optimal design is a balance between capital and operational cost as well as flexibility in the operation. Note that some of the lines are not plotted for all the time horizon as the simulation was terminated when the change in the variables was less than 5%.

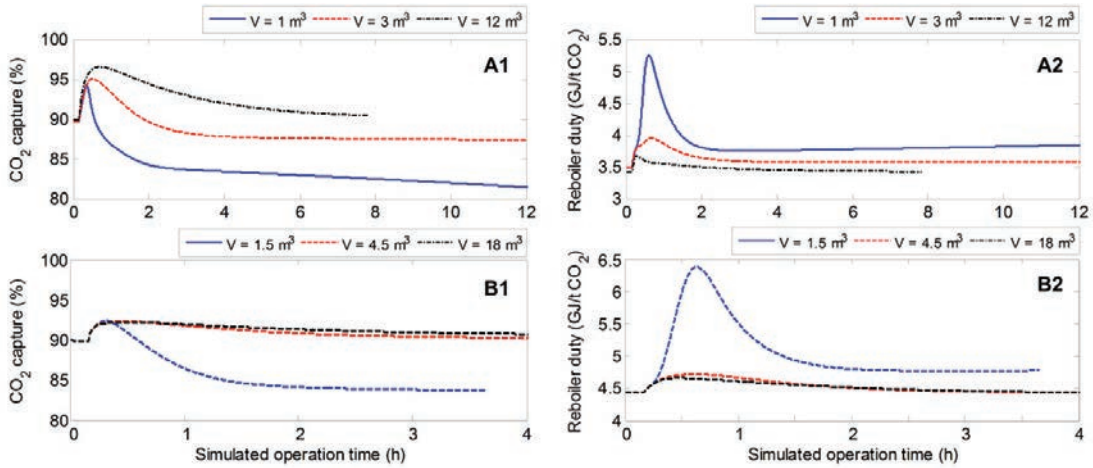

Fig. 3. Dynamic response of the plant for 10% step increase in the lean solvent flow rate using: (A1) and (A2) PZ; (B1) and (B2) MEA.

The steam supplied to the reboiler is another important manipulated variable in the control of a capture plant as power plants are required to accommodate sudden changes in the electricity grid. A possible approach to suddenly increase the electricity output of a power plant with integrated CO_2 capture is to reduce the amount of steam supplied to the reboiler unit, which leads to a constrain in the heat supply for the solvent regeneration process. Fig. 4 shows the effect of 10% step-decrease in the steam flow rate on the transient evolution of the PZ plant. The simulations showed that the dynamics of the plant is similar between PZ and MEA; thus, only the results for PZ are presented here for brevity.

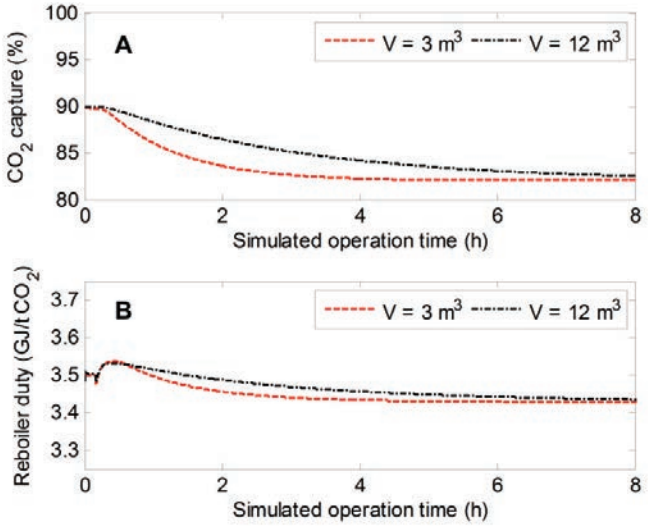

Fig. 4. Dynamic response of the plant for 10% step decrease in the steam solvent flow rate using PZ

Fig. 4A illustrates how the CO_2 capture percentage decreases when the steam supply is step-decreased. This change in the capture rate is faster and slightly greater for a smaller buffer tank. This lower capture rate is a result of the accumulation of CO_2 in the buffer tank, i.e. less CO_2 is stripped out in the desorber. The faster decrease of the capture efficiency in Fig. 4A is due to the accelerated buildup of CO_2 in the tank with 3 m^3 compared to the tank with 12 m^3, as expected. By inspecting the transient evolution in Fig. 4B, it can be observed that the volume of the tank does not influence significantly the reboiler duty. A shortage in the steam supply leads to a small initial increase in the heat demand followed by a slow decrease. After approximately 2 hours of operation, the reboiler duty is smaller than the nominal value, 3.5 GJ/t CO_2 for both buffer tank capacities. Thus, the sizing of the buffer tank will not influence significantly the operational cost of the capture unit with respect to changes in the steam flow but an under-designed buffer tank may lead to greater fluctuations in the controlled variables; therefore, a small tank results in peaks in the operational cost with respect to changes in the lean solvent flow rate.

These results suggest that the volume of the buffer tank plays a major role in the transient evolution of CO_2 capture rate and of the specific heat demand and the plant presents different responses when using buffer tanks of different capacities.

3.2. Parameter uncertainty in dynamic operation

This section presents a study that explores the relevance of uncertainties in the kinetic model on the performance of a PZ respectively MEA capture plant in the presence of 10% step-increase in the flue gas flow rate. The uncertainty of the kinetic model is an important aspect to consider, especially for novel and less-known solvents, e.g. PZ, enzyme enhanced MDEA, blends of PZ, etc. This scenario corresponds to ±10% uncertainty of the calculated mass transfer enhancement. Practically, the default enhancement factor (the value given by the GM enhancement factor model [11,12]) was changed by ±10% in the presence of 10% increase of the flue gas rate. The rest of the model parameters were kept constant.

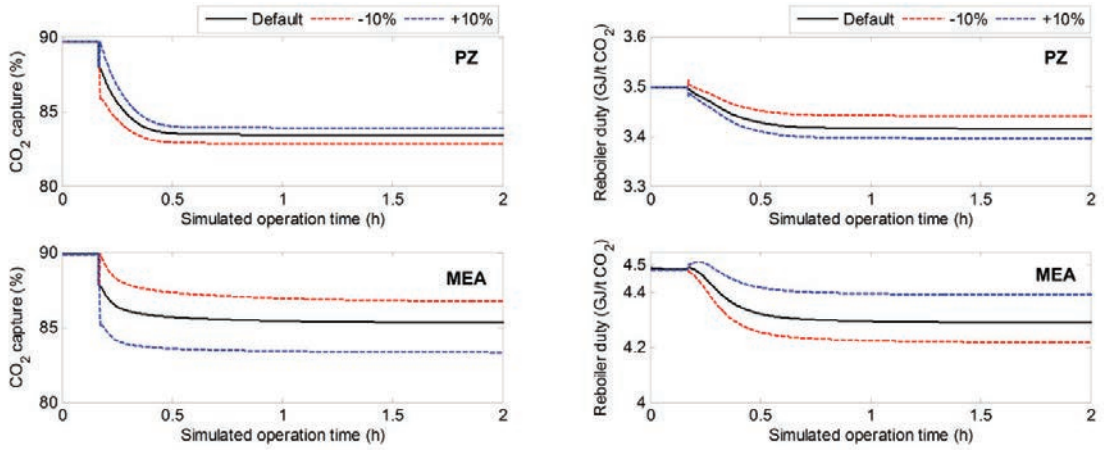

Fig. 5. Dynamic response of the plant for a 10% change in the flue gas flow rate with an uncertainty range of 10% in the kinetic model.

Fig. 5 shows the expected accuracy of the predicted CO_2 capture percentage and calculated reboiler duty for a 10% accuracy range of the kinetic model. The expected variability of these calculated properties corresponds to the range between the dashed red line and the dashed blue line. This figure illustrates how the accuracy of the kinetic model is more important for MEA compared to PZ model. Furthermore, Fig. 5 illustrates the differences between the dynamics of plants for the default case and the ±10% cases. In other words, the settling times and the gains of the process are different between the "default", "+10%" and "-10%" scenarios. The difference in the process gain is even more visible. The change in the PZ plant's performance with respect to change in the flue gas flow rate varies between 5.7% and 6.8% (CO_2 capture percentage) respectively 0.06 GJ/t CO_2 and 0.1 GJ/t CO_2 (reboiler duty). The change in the MEA plant's performance for 10% step change in the flue gas flow is between 3.2% and 6.6% (CO_2 capture percentage) and 0.09 GJ/t CO_2 and 0.26 GJ/t CO_2 (reboiler duty). Thus, Fig. 5 demonstrates that uncertainties related to the kinetic parameters have a great impact on the dynamics of the plant for both solvent, especially for MEA. This is important to remember when developing model based control or identifying a linear model. As a continuation of this study, the effect of uncertainties in other variables such as the hydraulic model, mass transfer model, thermodynamic model etc. will also be explored.

4. Conclusions

This work presented a comparison between the dynamics of a PZ and MEA plant for different capacities of the buffer tank and it showed the importance of design on economics and flexibility. Additionally, it exemplified the effect of uncertainties in the kinetic model on the transient evolution of the plant and the accuracy of the model. The results were created in Matlab using the dCAPCO2 dynamic rate-based model for CO_2 absorption and desorption.

This analysis showed that the volume of the buffer tank plays a significant role in the dynamics of the PZ and MEA plant for changes in the lean solvent flow rate and steam flow rate but the volume of the tank does not influence the transient evolution of the plant for changes in the flue gas flow rate. Furthermore, this study also showed that a relatively small buffer tank may lead to operational/controllability issues. The changes in the removal efficiency and reboiler duty are larger for the smallest studied tank capacity considered here. Furthermore, the PZ and MEA plant presents well-visible inverse responses in the capture rate for the smallest volume. These systems are challenging to control.

This preliminary study outlines the need for simultaneous design and controllability in order to develop a flexible and economically efficient post-combustion unit. The results presented here have illustrated that process dynamics and therefore the flexibility of the plant, and not only the steady-state behavior, has to be accounted for when designing a capture plant.

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