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Control of a post-combustion CO_2 capture plant during process start-up and load variations

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Abstract: Dynamic and flexible operation of a carbon capture plant is important as thermal power plants must be operated very flexibly to accommodate large shares of intermittent energy sources such as wind and solar energy. To facilitate such operation, dynamic models for simulation, optimization and control system design are crucial. In this paper, we present a dynamic mathematical model for the absorption and desorption columns in a carbon capture plant. Moreover, we implement a decentralized proportional-integral (PI) based control scheme and we evaluate the performance of the control structure for various operational procedures, e.g. start-up, load changes, noise on the flue gas flow rate and composition. Note that the carbon capture plant is based on the solvent storage configuration. To the authors knowledge, this is the first paper addressing the issue of start-up operation and control of carbon capture. The study demonstrates that the implemented control structure keeps the carbon capture process at 90% CO_2 removal rate with a deviation up to 8% during load variations. In addition, it reveals that the control structure brings the process to the desired set point in approximately 10 min during process start-up.

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Keywords: Process Applications; Energy Processes and Control; Modeling and Identification

1. INTRODUCTION

Carbon Capture and Storage (CCS) has witnessed high growth in recent years due to increasing climate and environmental concerns, as well as regulatory norms established by governments. The role of CCS in sustainable energy infrastructure development is becoming unanimously accepted. The growing focus to reduce CO_2 emission imposes the need to implement CO_2 capture in fossil-fuel fired thermal power plants. However, issues related to energy intensiveness of CCS technologies and flexibility of the power plant with integrated CO_2 capture need to be addressed. For example, power plants are required to have the capacity to increase their energy output by 10% over a period of 10 s, according to the UK Grid code. Moreover, power plants need to be able to balance the changes in the energy demand within a 30 sec to 30 min period (Lawal et al. (2010)). Such a flexibility demand on the thermal power plants requires that the carbon capture plant also can be operated in a very flexible manner.

Power plants with integrated carbon capture plants have to be optimized and controlled to enable flexible and economically efficient operation. Development of control structures and model based optimization techniques require dynamic models of the individual units and of the entire plant. The main units of a CO_2 post-combustion capture (PCC) plants are the packed columns for CO_2 absorption and desorption. Dynamic models for the simulation of the absorber and desorber have recently started to appear in the open literature. The complexity of these models depends on the approach used to describe mass transfer and chemical reaction. The simplest model assumes vapour-liquid and chemical equilibrium. The most complex model takes into account the vapor-liquid phase change, the mass transfer between the phases and the reaction kinetics between the species, as shown by Biliyok et al. (2012). In practice, existing dynamic models balance between accuracy and complexity to keep control of the computational load. Lawal et al. (2010) presents a dynamic model in gPROMS with a rate-based model formulation and assumes chemical equilibrium. Kvamsdal et al. (2009), Gaspar and Cormos (2011), and Harun et al. (2013) also use the rate based formulation with the enhancement factor approach to account for the reaction kinetics. However, they use constant values for some mass transfer and hydraulic properties, such as hold-up, wetted area, and pressure drop. In addition, these models are based on simplified thermodynamic models. In the present paper, we also describe the absorber and stripper with a rate-based model using an enhancement factor for the calculation of the mass transfer rate. However, the developed model accounts for changes of the mass transfer and hydraulic characteristics along the column height using the Rocha et al. (1993, 1996) model. The extended UNIQUAC thermodynamic model by Thomsen and Rasmussen (1999) describes the vapor-liquid phase change and thermal properties.

The control and optimal operation of carbon capture processes receive more and more attention with the deployment of the first industrial scale post-combustion capture plant at the Boundary Dam coal-fired power plant. Lin et al. (2011) demonstrates that the amine solution flow rate, the lean loading, and water make-up have to be maintained to achieve acceptable closed-loop process performance. They propose to control the CO_2 removal rate by manipulating the lean solvent flow rate to the absorber. In addition, they select the steam flow to the reboiler to regulate the lean loading of the solvent. They evaluate the performance of the control scheme for step changes up to 20% of various process parameters. Nittaya et al. (2014) and Sahraei and Ricardez-Sandoval (2014) present a control

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structure to maintain the 90% CO_2 removal rate and to keep the CO_2 product stream purity above 95%. Nittaya et al. (2014) presents control schemes based on Relative Gain Array (RGA) and heuristic approaches, while Sahraei and Ricardez-Sandoval (2014) explore the benefits and limitations of a multivariable model-based control technique. Both, Nittaya et al. (2014) and Sahraei and Ricardez-Sandoval (2014), focus on studying small changes of the flue gas flow rate. However, they investigate the response of the control configuration for constrained reboiler heat duty. Although, these works show the performance of the developed control structure in the presence of various step and ramp changes, none of the works assess the performance of the control scheme for periods of start-up, shut down and large changes of the power plant load.

The scope of this article is to explore the benefits and limitations of a decentralized control technique to address the flexibility of a post-combustion carbon capture plant. We implement two SISO controllers to keep the CO_2 removal rate and the lean loading at the desired set points. The focus is on the performance evaluation of the absorber. To achieve this, we investigate the dynamic open-loop and closed-loop performance of the carbon capture plant during start-up and large load changes of the power plant. Moreover, this paper presents a mechanistic first principle based dynamic model for transient simulation of packed columns, e.g. CO_2 absorber and desorber. The model uses heat and mass transfer correlations and it is based on rigorous thermodynamics. The present modeling and controllability analysis is carried out in Matlab.

The structure of the paper is as follows: Section 2 describes the details of the mechanistic first principle based CO_2 capture dynamic model. Section 3 continues with a brief overview of the process boundaries. Section 4 proposes a design for the control of the carbon capture plant and describes a step response study to gain knowledge about the dynamics of the system. Section 5 demonstrates the flexibility of the carbon capture plant with solvent storage configuration by simulating periods of start-up and large load changes of the carbon capture plant. The paper summarizes the findings in the Conclusions section.

2. DYNAMIC MODEL DEVELOPMENT

In this section, we provide an overview of a mechanistic first principle based model for CO_2 absorption and desorption using monoethanolamine (MEA). The mathematical model of the absorber and stripper are similar with the exception that the stripper requires a reboiler unit. Mass and energy balances for the gas and liquid phase represent the base of the model. These equations depend on heat and mass transfer fluxes between the liquid phase and the gas phase and on the rate of reaction between CO_2 and MEA.

To keep the model flexible and generic, we consider that the mass transfer between the phases is bi-directional. Positive sign shows mass transfer from gas to liquid, i.e. absorption or condensation. Negative sign refers to desorption and evaporation. In addition, we assume:

- The flow fields of the column are strongly turbulent.
- No accumulation in the gas and liquid films.
- MEA is non-volatile.
- Reaction takes place only in the liquid film.
- Heat loss to the surroundings is negligible.

2.1 Mass and energy conservation equations

The plug-flow model is a common model to describe packed columns, e.g. the CO_2 absorber and desorber. The conservation of mass and energy is applied to a control volume of height Δz. The liquid inlet concentration, temperature and flow rate are fixed at the top of the column. The gas inlet conditions are fixed at the bottom of the column. The liquid bottom and gas top streams are determined using the conservation laws. The resulting differential mass balances for component i in the gas phase and the liquid phase are as follow:

$$\frac{\partial y_i}{\partial t} = -\frac{1}{C_g}\frac{\partial N_{i,g}}{\partial z} - \frac{a_{pck}}{\epsilon(1-h_l)}J_{i,gl} \tag{1}$$

$$\frac{\partial X_i}{\partial t} = \frac{1}{C_l}\frac{\partial N_{i,l}}{\partial z} + \frac{a_{pck}}{\epsilon h_l}J_{i,gl} \tag{2}$$

y_i is the mole fraction of component i in the gas phase; X_i is the apparent mole fraction of component i in the liquid phase. The apparent mole fraction expresses the composition of the liquid on a CO_2-free basis. C_g and C_l are the molar density of the gas and liquid phase (mol/m^3); $N_{i,g}$ and $N_{i,l}$ denotes the effective gas and liquid flux of component i (mol/m^{2}s); $J_{i,gl}$ is the mass transfer flux of component i through the gas-liquid interface. Component i can be CO_2, H_2O and MEA. ϵ is the void fraction, h_l is the liquid hold-up of the column and a_{pck} is the effective gas-liquid contact surface.

To facilitate the application of the energy conservation principles, we assume that the mass and heat transfer area are equal. In addition, it is assumed that the volatile components condense at the gas-liquid interface releasing the heat to the liquid. Based on these assumptions, the conservation of energy for the gas phase and the liquid phase are

$$\frac{\partial T_g}{\partial t} = -\frac{1}{C_g c_{p,g}}\frac{\partial Q_g}{\partial z} - \frac{a_{pck}}{\epsilon(1-h_l)C_g c_{p,g}}q_{cond} \tag{3}$$

$$\frac{\partial T_l}{\partial t} = \frac{1}{C_l c_{p,l}}\frac{\partial Q_l}{\partial z} + \frac{a_{pck}}{\epsilon h_l C_l c_{p,l}}(q_{conv} + q_{cond} + q_{gen}) \tag{4}$$

where T_g and T_l are the temperature of the gas and liquid phase; Q_g and Q_l is the effective heat flux (J/m^{2}mol); $c_{p,g}$ and $c_{p,l}$ denote the specific heat capacity of the gas respectively liquid phase. The heat transfer between the gas and liquid phase due to temperature difference is

$$q_{cond} \approx \lambda(T_g - T_l) \tag{5}$$

In eq. (5), the Chilton-Colburn analogy describes the heat transfer coefficient, λ. The convective heat transported through the gas-liquid interface, q_{conv}, by the volatile components, i.e. CO_2, H_2O is:

$$q_{conv} = T_g \sum J_{i,gl} c_{p,i} \tag{6}$$

Eq. (7) includes the heat of physical absorption and chemical reaction, known as heat of absorption ($\triangle_r H$), plus the heat of vaporization ($\triangle_v H$). They are calculated with the extended UNIQUAC thermodynamic model (Thomsen et al., 1996).

$$q_{gen} = J_{CO_2,gl} \triangle_r H + J_{H_2O,gl} \triangle_v H \tag{7}$$

Although, the mass transfer and hydraulic characteristics may be relatively constant at steady-state, at start-up, shut down, and

at large load changes, these parameters vary significantly. To account for these parameters, the present work implements the model of Rocha et al. (1993, 1996).

2.2 Simultaneous mass transfer and reaction model

The two-film model, the penetration model, or the surface renewal model can be used to describe the mass transfer phenomena of reacting systems. However, the solution of these models is computationally demanding. In process simulation, the so called enhancement factor approach is preferred, since it reduces the computational time and complexity of the numerical scheme. The enhancement factor gives an approximate solution of the mentioned transfer models. It is defined as the ratio of mass transfer from chemical absorption compared to physical absorption. The present model is intended in a two-film approach and it is based on the General Method (GM) model, presented in Gaspar et al. (2014a,b). Therefore, the CO_2 and H_2O transfer rates through the gas-liquid interface, $J_{i,gl}$, are

$$J_{i,gl} = -D_i \frac{\partial C_{i,g}}{\partial x}|_{x=0} \approx K_{i,g}^{tot}(p_i^g - p_i^{eq}) \quad (8)$$

$$\frac{1}{K_{i,g}^{tot}} = \frac{1}{k_{i,g}} + \frac{H_i}{E k_{i,l}} \quad (9)$$

p_i^g denotes the partial pressure of component i in the gas phase and p_i^{eq} is the equilibrium partial pressure of component i. Eq. (9) describes the total mass transfer coefficient, $K_{i,g}^{tot}$, where $k_{i,g}$ and $k_{i,l}$ are the partial mass transfer coefficients in the gas and liquid film. E is the enhancement factor and H_i is the Henry coefficient for component i.

2.3 Thermodynamic model

The model for CO_2 absorption and desorption uses the extended UNIQUAC model for the determination of the vapor-liquid equilibrium and thermal properties. The extended UNIQUAC is a Gibbs excess model which applies the thermodynamic γ-ϕ approach. Therefore, liquid phase activity coefficients are calculated with the extended UNIQUAC model, and the gas phase fugacity coefficients are estimated with the Soave-Redlick-Kwong equation of state. The extended UNIQUAC model is a combination of the original UNIQUAC model and a Debye-Hückel term. The original UNIQUAC model consist of the combinatorial and residual terms (Abrams and Prausnitz (1975)). The Debye-Hückel term corrects for the electrostatic interactions between the ions in the liquid phase, and it is

$$\frac{G^{DH}}{RT} = -x_w M_w \frac{4A}{b^3}(ln(1 + b\sqrt{I}) - b\sqrt{I} + \frac{b^2 I}{2}) \quad (10)$$

where x_w and M_w are the mol fraction and molecular weight of water; I is the molal ionic strength of the compounds in mol/kg H_2O and b denotes the molal concentration. Note that A is a temperature dependent adjustable parameter. Thomsen et al. (1996) outline the details of the equations system and derived properties. Thomsen and Rasmussen (1999) show the method for performing the VLSE calculations and Thomsen (2005) details how parameter fitting is performed and outlines a few possibilities for use of the model.

Table 1. Base case process design and inlet parameter specifications

Flue gas flow rate	9.64 kmol/s
Flue gas inlet temperature	43.5^{o}C
Flue gas inlet CO_2 mol fraction	14.1%
Lean solvent flow rate	39.02 kmol/s
Lean solvent inlet temperature	40 ^{o}C
MEA concentration of the solvent	30 wt.%
Lean solvent loading	0.21 mol/mol
Stripper's feed temperature	98 ^{o}C
CO_2 removal rate	90%
Height of the absorber/stripper	18 and 12 m
Diameter of the absorber/stripper	10 and 8 m

3. BOUNDARIES OF THE PROCESS

Fig. 1 shows the process flow-sheet with the implemented control scheme. The base for the design of the capture plant is the traditional process configuration with heat integration between the lean solution leaving the reboiler and the rich solution entering the stripper. In addition, we consider the option of storing a fraction of the lean solvent in a buffer tank and the rich solvent in a storage tank during start-up, periods of peak electricity prices and shut down operations. The role of the two tanks is to reject small disturbances in the solvent loading and to decouple the operation of the power and capture plant (Dowell and Shah, 2014). Moreover, these tanks partially decouple the operation of the absorber and desorber and they increase the flexibility of the CO_2 capture plant. Here, an extra parameter is the quantity of the solvent stored and/or regenerated in each time period. However, we focus on operation of the carbon capture plant for short periods of time and we do not address this scheduling issue. In this paper, we assume that the rich solvent flow rate to the stripper is constant and the capacity of the buffer tank is able to supply the required lean solvent flow to the absorber.

The base case operating conditions correspond to a 200 t/hr CO_2 capacity post-combustion capture plant using 30 wt.% MEA solution. The loading of the lean solution entering the absorber is approximately 0.21 mol CO_2/mol MEA at 40^oC. The flue gas comes from a coal-fired power plant. Therefore, it contains 14.1 mol% CO_2 and it is saturated with water before entering the absorber. In addition, we assume that the SO_x and NO_x content of the flue gas is removed upstream of the capture plant.The carbon capture plant is designed for 90% CO_2 removal rate and it is equipped with three heat exchangers. These heat exchangers secure the cooling of the lean solvent and provide constant preheating of the rich solution entering the stripper. Table 1 summarizes the main design specifications

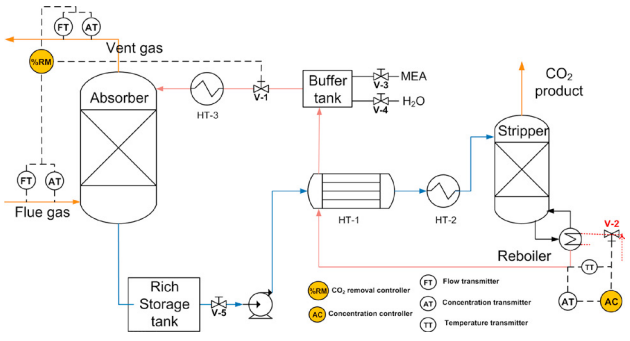

Fig. 1. Base case process flowsheet with control structure

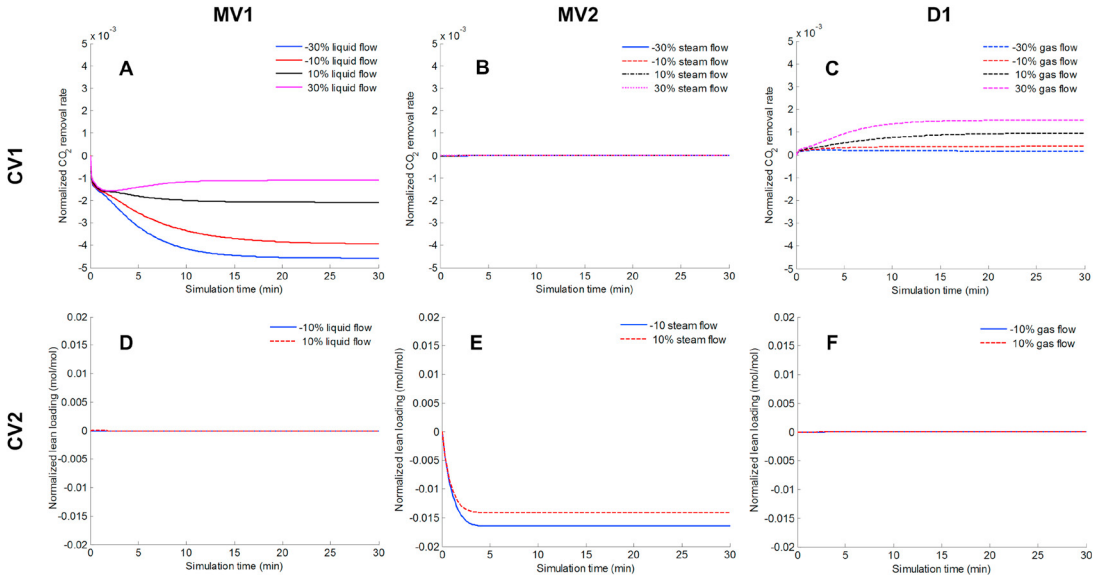

Fig. 2. Step response analysis of the carbon capture plant. (MV1 - lean solvent flow rate, MV2 - steam flow rate, D1 - flue gas flow rate, CV1 - CO_2 removal rate and CV2 - lean loading)

and process parameters for the post-combustion CO_2 capture plant.

The governing units of the post-combustion process are the columns for absorption and desorption. In this study, we neglect the dynamics of additional units, i.e. the pump, the heat exchangers, the buffer tank to simplify the solution of the problem. This is a reasonable assumption for the partially decoupled absorber and desorber, i.e. configuration with storage buffer tank.

4. DESIGN OF THE CONTROL STRUCTURE

This section briefly outlines the design of the control structure proposed for the CO_2 capture process described in the previous section. It relies on the work of Nittaya et al. (2014) and Sahraei and Ricardez-Sandoval (2014). They develop and implement a plant-wide control structure based on a decentralized multi-loop control scheme and a model-based predictive control technique. Although, they evaluate the controllability of the CO_2 capture process for sudden and sustained changes in the plant's load, the behavior of the carbon capture plant for start-up and large load decrease is not investigated. To the author's knowledge, this is the first work addressing the control of a post-combustion capture plant for a broad range of operating conditions, i.e. start-up, and large load changes.

4.1 Control objectives and process variables

Similar to Nittaya et al. (2014) and Sahraei and Ricardez-Sandoval (2014), the control objective is to bring and keep the process at 90% CO_2 removal as well as to keep the composition of the lean solution at 0.21 CO_2 loading. Fig. 1 outlines the implemented control structure for the carbon capture plant. It shows that the selected control variables for this system are the CO_2 removal rate (CV1) and the lean loading out of the reboiler (CV2).

To maintain the process at the desired set points, the flow rate of the lean solution to the absorber (MV1) and the flow rate of the steam to the reboiler (MV2) are manipulated. The steam flow determines the temperature of the reboiler and influences the loading of the solution. Similar to Panahi and Skogestad (2012), the control of the water and MEA make-up flow is manual. The purity of the CO_2 product stream is determined by the condenser operating temperature. Therefore, its control can be decoupled form the capture plant operation and it is not considered in the present work. In this work, we implement two SISO control structures: a proportional-integral controller to keep the removal rate and a proportional controller to adjust the lean loading. For the tuning of the controllers, we apply the SIMC-PID tuning method by Skogestad (2003). This is a step response based procedure with one tuning parameter.

4.2 Step response study

The next step is to perform a step response study. For this, a disturbance of ± 10 to $\pm 30\%$ is applied to the lean solvent, flue gas and reboiler steam flow rate. This analysis contributes to the understanding of the dynamics of the process and it allows determining the process gains and time constants, needed for tuning of the controllers.

Fig. 2 shows the normalized step responses. Fig. 2A to Fig. 2C illustrate the correlation between the CO_2 removal rate, CV1, and lean solvent (MV1), flue gas (D1) and reboiler steam flow rate (MV2). Fig. 2D to Fig. 2F substantiates the correlation between the lean loading, CV2, and (MV1), (MV2), and (D1). The step response analysis substantiates the direct correlation between the CO_2 removal rate and the lean solvent flow. Furthermore, it outlines the inverse correlation between the removal rate and the flue gas flow rate. Fig. 2B underlines that CV1 does not depend on MV2 due to the decoupling of the units. Fig. 2D and Fig. 2F demonstrate that the lean loading, CV2, is independent of the solvent flow rate, MV1 and the flue gas flow rate, D1.

5. RESULTS AND DISCUSSION

In this section, we discuss the performance of the implemented control structure. It illustrates the behavior of the con-

trol scheme for various operational scenarios, i.e. start-up operation, large increase and decrease of the load of the carbon capture plant. Fig. 3, Fig. 4 and 5 show the simulation results.

In practice, various disturbances appear in a complex process such as power plant operation with integrated carbon capture. One of the most important disturbances represent the variation of the flue gas flow rate and composition. Note that the flue gas is cooled-down in direct contact coolers before entering the absorber. Therefore, its water and CO_2 content may vary. We mimic these oscillations by applying a normally distributed pseudo-random noise to the CO_2 content and the flue gas flow rate.

5.1 Start-up operation

This scenario presents the performance of the implemented controller during start-up. There are two possibilities to start a carbon capture plant: (1) the flue gas enters the absorber after steady state is reached with air and solvent; (2) the flue gas is lead through the absorber upon start-up and the amine solvent flow is adjusted accordingly to remove 90% CO_2. The second approach minimizes the CO_2 emission.

In the present work, we implement and simulate approach (2). It should be emphasized that many of the correlations for mass transfer coefficients and properties contain dimensional groups raised to non-integer powers. If these groups become negative during the simulation, the solver will fail. As the start-up condition implies model variables at or near zero, these conditions might arise.To avoid this issue, we assume that the column is pressurized with ambient air and that the initial solvent flow rate is 20% of the steady-state value.

Fig. 3 shows the performance of the controller for approach (2). In addition, it illustrate the behavior of the process when the liquid to gas (L/G) ratio is kept constant at its steady state optimal value. This figure also shows the manipulated variable (liquid flow rate) and the flue gas flow rate as function of time. Fig. 3 illustrates that the PI controller performs better than a controller which keeps a constant (L/G) ratio. The PI controller reaches the set point in approximately 10 min, while the constant (L/G) ratio controller reduces the removal rate to the set point in approximately 20 min. Therefore, the PI controller reduces faster the CO_2 removal rate and it keeps at 90% with an accuracy of $\pm 3.5\%$.

5.2 Load changes of the power plant

This scenario describes the performance of the CO_2 capture plant for large and sudden changes of the flue gas flow rate. This is a typical scenario. Coal-fired thermal power plants need to be operated very flexibly to accommodate large shares of intermittent energy sources, such as wind and solar energy.

In this study, we introduce combined step and ramp changes of -50% and 30% in the flue gas flow rate with respect to its nominal, full load operating point. To achieve this, we apply a step decrease of 10% after 1 hour of steady-state operation. Then, we reduce the flue gas flow rate to 50% in 2 minutes. After 2 hours, the flue gas flow rate is ramped up to 130% of its steady-state value. Fig. 4 and Fig. 5 show the behavior of the control structure for this scenario. It also presents the open-loop response of the process, to highlight the performance of the controller.

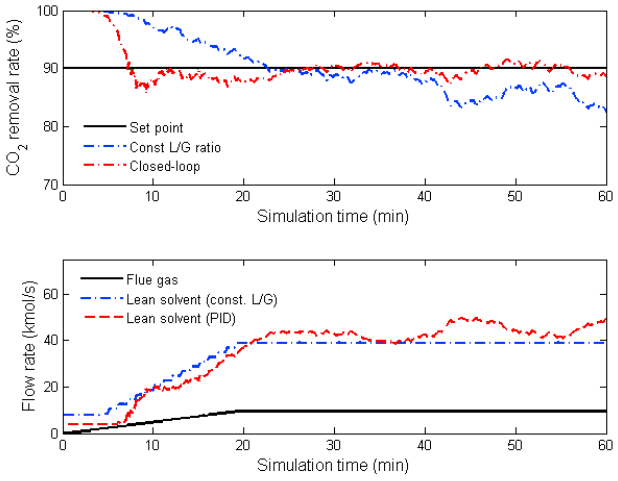

Fig. 3. Performance of the PID and ratio controller during start-up operation.

Fig. 4 substantiates that a small spike appears in the CO_2 removal rate when a disturbance appears. However, the PI-controller for the CO_2 removal rate adjusts the liquid flow rate and brings the process to the set point in $7 - 10$ min. It can be seen that the controller regulates the liquid flow rate and keeps the process at 90% CO_2 removal rate. Without feedback-control (open-loop), the removal rate first would increase to 100% than would decrease to approximately 75%. Although, the rich storage tank flattens the effect of the disturbance on the stripper, small changes of the CO_2 loading of the feed to the stripper may appear. However, the second control loop for the lean loading corrects for these disturbances on the rich solvent. Fig. 5 shows that the PI-controller increases the steam flow rate when the inlet rich loading increases and it reduces the steam flow when the rich loading drops.

Fig. 4 and Fig. 5 underlines the good performance of the control structure. The deviation from the set point is generally $\pm 1\%$. Even more, the controller manages to keep the CO_2 removal rate between $90 \pm 8\%$ all along the simulation and the loading of the solution at 0.21 ± 0.02. Therefore, the proposed PI-controller rejects large load changes of 50% and 130%.

6. CONCLUSIONS

In this paper, we present a dynamic mathematical model for the absorption and desorption column in a carbon capture plant. We implement and analysis a decentralized proportional-integral (PI) based controller to keep the carbon capture plant at 90% CO_2 removal rate and 0.21 mol CO_2/mol MEA lean loading. We evaluate the performance of the implemented control structure during process start-up and large load variations.

The analysis reveals that the PI-controllers are able to keep the plant at the desired set-point with $\pm 8\%$ deviation. Note that the simulations are based on a 200 t/hr capacity post combustion plant using monoethanolamine. The base for the process configuration is the solvent storage approach which allows decoupling of the power and capture plant. Moreover, it partially decouples the operation of the absorber and desorber and it increases the flexibility of the carbon capture plant.

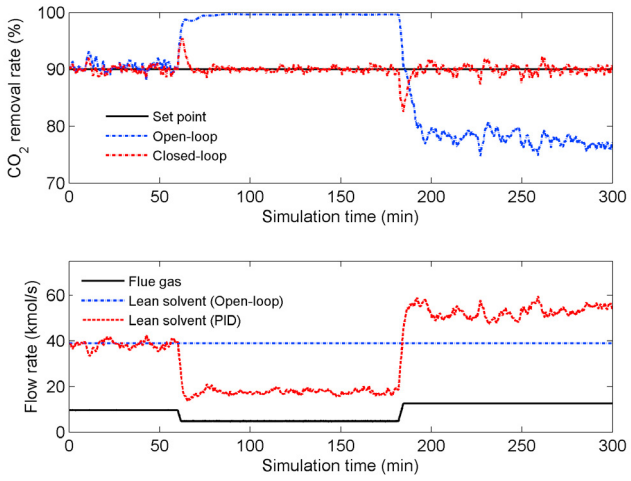

Fig. 4. Performance of the control structure for a 50% decrease and 30% increase of the load of the PCC.

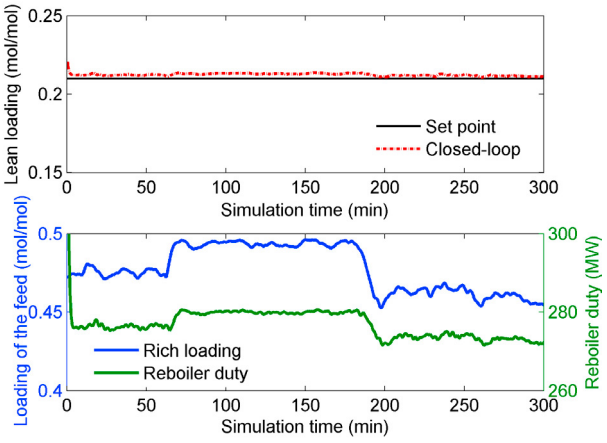

Fig. 5. Response of the lean loading controller during load changes of the PCC.

We aim to couple the dynamic model for CO_2 absorption and desorption with models of other sub-units (heat exchangers, sumps, pumps, buffer tanks) to form the complete model of a post-combustion capture plant. The carbon capture model is intended for development of model-based-control structures. In addition, advanced process configurations, such as lean-vapor-re-compression, solvent split flow, inter-cooling, etc. will be tested to evaluate their potential to reduce the energy intensiveness of the carbon capture process.

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