

This is a repository copy of Application of piece-wise linear system identification to solvent-based post-combustion carbon capture.

White Rose Research Online URL for this paper: http://eprints.whiterose.ac.uk/140071/

Version: Accepted Version

Article:

Liao, P., Wu, X., Li, Y. et al. (4 more authors) (2018) Application of piece-wise linear system identification to solvent-based post-combustion carbon capture. Fuel, 234. pp. 526-537. ISSN 0016-2361

https://doi.org/10.1016/j.fuel.2018.07.045

Article available under the terms of the CC-BY-NC-ND licence (https://creativecommons.org/licenses/by-nc-nd/4.0/).

Reuse

This article is distributed under the terms of the Creative Commons Attribution-NonCommercial-NoDerivs (CC BY-NC-ND) licence. This licence only allows you to download this work and share it with others as long as you credit the authors, but you can't change the article in any way or use it commercially. More information and the full terms of the licence here: https://creativecommons.org/licenses/

Takedown

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing eprints@whiterose.ac.uk including the URL of the record and the reason for the withdrawal request.



eprints@whiterose.ac.uk https://eprints.whiterose.ac.uk/

Application of piece-wise linear system identification to

solvent-based post-combustion carbon capture

Peizhi Liao^a, Xiao Wu^a, Yiguo Li^{a, *}, Meihong Wang^{b, *}, Jiong Shen^a, Adekola Lawal^c, Chuanlong Xu^a

^aKey laboratory of Energy Thermal Conversion and Control of Ministry of Education, Southeast University, Nanjing 210096, China ^bDepartment of Chemical and Biological Engineering, University of Sheffield, Sheffield S1 3JD, UK ^cProcess Systems Enterprise Ltd, 26-28 Hammersmith Grove, London W6 7HA, UK

Abstract

1

2

3

Solvent-based post-combustion carbon capture (PCC) is currently the most promising method to reduce CO₂ emission. To achieve a plant-wide controller for flexible operation, it is necessary to develop a data-driven model to understand the dynamic characteristics of PCC plant. This paper aims to: (i) carry out system identification to develop a data-driven model and (ii) provide insights into the nonlinear dynamics among the key variables from the PCC process in a wide operating range. These key variables include: CO₂ capture rate, reboiler temperature, condenser temperature and lean solvent temperature. Pilot-scale PCC process implemented in gCCS was used to generate simulation data for system identification and model comparison. Linear single-input-single-output (SISO) transfer function models were firstly developed at different capture rates. Open loop step tests on identified models were then introduced to report the dynamics of key variables in various operating conditions and to indicate the level of system nonlinearity graphically. The nonlinearity analysis was carried out to investigate the system nonlinearity distribution in a quantitative manner. Based on the nonlinearity analysis, a multi-input-multi-output (MIMO) piece-wise model was proposed to simulate the nonlinear characteristics of PCC plant. The piece-wise model shows a satisfactory agreement with gCCS simulation data. Results of this study successfully demonstrate the nonlinear behavior of the solvent-based PCC process, which can be applied in the design of flexible plant-wide controllers.

Keywords: Post-combustion carbon capture; Solvent-based carbon capture; System identification; Piece-wise model; Nonlinearity analysis

1. Introduction

1.1 Background

Global warming has increasingly drawn public attention. Extensive research has been performed to combat this trend. The Intergovernmental Panel on Climate Change (IPCC) stated that CO_2 contributed to about 50% of the increasing temperature in the earth surface among all the greenhouse gas [1]. Most of the CO_2 emissions originate from combustion of fossil fuels in large-scale power plants. For the near future, it is necessary to take measures to reduce CO_2 emission from these sources since fossil fuel is still attractive to meet future energy demands due to its rich availability, large energy density and low cost [1].

Among all the approaches, solvent-based PCC technology is viewed as the most mature option for existing power plants [3]. It offers advantages over other capture technologies because of high selectivity and pure CO₂ stream collection [3]. It can also be retrofitted as an end-of-pipe solution.

In the past decades, research efforts have been devoted to understanding the intricate nature of this carbon capture process. Solvent regeneration is energy intensive and it requires a lot of steam extracted from power plants. Given its high energy requirement, the solvent regeneration process will reduce the overall power plant efficiency significantly [4]. It is therefore important to minimize the energy demand and make more steam available for power generation [5]. However, it is still a matter of concern to find a trade-off to balance CO_2 removal rate and energy cost under the time-varying economic conditions. In this regard, there is a need to develop a flexible plant-wide control structure in order to achieve optimal performance in the presence of disturbance, load-changing and other scenarios. The nonlinear dynamic characteristics of PCC process need to be analyzed to provide information for the advanced controller's design.

42 1.2 Motivation

To study the dynamics of solvent-based PCC plant, several first-principle models have been developed (Lawal et al [5, 6, 7]). These models have been proven to be able to closely predict the real process. A number of carbon capture pilot plants are now 45 available worldwide to provide steady-state or dynamic validation data (Dugas [8], Biliyok et al [9]). Nevertheless, it is clear that 46 the simulation with first-principle models is computationally demanding. This makes controller design based on first-principle 47 models difficult. Therefore, it is necessary to use a data-driven black-box identification method to serve as an alternative. On the other hand, most of the studies for PCC plant are focused on the linear models developed at a fixed operating point. Under varying 48 operating conditions, the transient behavior of PCC plant will change and it would be difficult for linear models to simulate the 49 50 nonlinear features. The linear model's failure to capture the nonlinear dynamics of the PCC plant will deteriorate the control 51 performance. In order to develop a model predictive controller for wide-range capture rate change, Wu et al [10] proposed a simple 52 nonlinear distribution analysis for solvent-based PCC plant. However, since multi-variable model is used in the analysis, the 53 nonlinearity for a certain input-output loop cannot be revealed in detail. Motivated by these shortcomings, an investigation was 54 carried out to further understand the operational features of a PCC plant over a wide range of operating conditions, and to identify 55 possible control difficulties which may arise.

56 1.3 Aim of the study and main novel contributions

57 This study aims to identify a plant-wide black-box model based on piece-wise linear identification method. There are two major 58 novelties in this paper:

- A fuzzy-based piece-wise model which can approximate the nonlinear dynamic features of a PCC process from 50% 95%
 capture rate is achieved at a fixed power plant load. This model can be used for the plant-wide controller design.
- Detailed nonlinear characteristics of key variables in PCC process are researched quantitatively. These key variables include:
 CO₂ capture rate, reboiler temperature, condenser temperature and lean solvent temperature.

63 1.4 *Outline*

The paper is organized as follows: Section 2 presents the available literature review on modelling and identification of PCC process. Section 3 generates the simulation data of solvent-based PCC process in gCCS platform and uses these data to identify linear local SISO system models from 50% to 95% capture rate. In Section 4, a critical sensitivity analysis is performed by introducing step changes in the input variables. Nonlinearity degree analysis is carried out in Section 5. Section 6 presents the MIMO fuzzy-based piece-wise model. Conclusions are drawn in Section 7.

69 **2. Literature review**

Mass transfer and chemical reaction are two key factors to consider in modelling solvent-based PCC process. To describe the mass transfer process, two approaches are usually used in most studies: the equilibrium-based approach and the rate-based approach. In Lawal et al [6], a critical comparative evaluation showed that a rate-based model gives better agreement with experimental data.

To date, many studies on dynamic modelling have been implemented. In Kvanstal et al [11], a dynamic model of standalone absorber column in rate-based modelling approach was presented. This model was simulated in two load-varying cases, namely, start-up and load-reduction, to evaluate the operability of absorber. In Ziaii et al [12], a standalone stripper model was built in Aspen Customer modelling environment. Dynamic simulation was carried out to run the stripper flexibly during the period of high electricity demand and price.

However, the limitation of the aforementioned publications is that stand-alone model cannot represent the whole PCC process 78 79 due to the intricate nature with regard to high nonlinearity and process interactions. Therefore, a dynamic model considered 80 interacted units is significant to combine them together as a whole plant. Lawal et al [5] presented a dynamic model including absorber, stripper and recycle. Based on a comparative assessment, the whole process model gives more accurate results in 81 82 predicting temperature profile than standalone columns. In their follow-up work [7], a scaled-up integrated model to industrial size 83 of a 500 MW coal-fired subcritical power plant was made available. This work gave a preliminary technical evaluation of integrated 84 PCC process and power plant. Due to lack of experimental data, dynamic validation is very rare. Biliyok et al [9] presented data at 85 transient scenarios for dynamic model validation. In the same paper, dynamic process analysis proved that mass transfer is the 86 major factor which limits CO₂ absorption.

87 First-principle model provides the advantage to realize accurate simulation, as well as understanding the underlying dynamics

- 88 of the process. Nevertheless, as stated previously, first-principle model is computational intensive and it is hard to realize. Thus, 89 carrying out black-box identification has emerged as an attractive alternative to first-principle dynamic modelling.
- Arce et al [13] used MatlabTM identification toolbox (Ljung [14]) to obtain a linear model for solvent regeneration process. This model was composed of a first-order linear discrete transfer function with a sampling time of 200 ms. However, a first-order model cannot mimic the dynamic features compared with a higher order model. For capturing nonlinear characteristics, Manaf et al [15] employed a multivariable nonlinear autoregressive with exogenous input (NARX) model. To reduce computational demand, identified model for absorber, rich/lean heat exchanger and stripper were acquired separately and united as a 4-input-3-output PCC model. The distinguished contribution of [15] is that the rich/lean heat exchanger, a major investment and operating penalty unit [16], was considered in the modelling. However, this paper did not provide available control structure to operate heat exchanger.
- 97 Li et al [17] presented a bootstrap aggregated neural approach to build a multiple-inputs-single-output (MISO) dynamic model. 98 Results showed its superiority in predicting capture level compared with conventional neural networks. One-step-ahead and multi-99 step-ahead prediction were used as the neural network input. It was found that one-step-ahead prediction is more accurate, because 100 the prediction errors were accumulated every sampling time in a multi-step-head prediction and this would increase the prediction 101 error at the following sampling time.
- In data-driven modelling of a nonlinear system, the most typical way is to use polynomial functions to approximate the nonlinearity [18]. But the model order resulted from the polynomial function is always high, especially for a complicated industrial process. It is not always easy to solve these equations analytically. There is another option for nonlinear approximation which uses piece-wise-linear (PL) functions. PL functions aim to approximate the nonlinear features by a combination of linear pieces. In general, the nonlinear model produced by a serial of linear models is expected to result in an easier implementation, theoretical analysis and calculation [19]. The proposed work tries to use a piece-wise modelling method to simulate the real PCC process, and also to investigate the nonlinear features.

109 **3. Process description and local model identification**

110 3.1. Process description

gCCS simulation software was developed based on gPROMS modelling platform to support simulation and design of power plants, carbon capture, transport and storage [20]. It was developed by Process Systems Enterprise (PSE) Ltd in London and it is commercially available. As shown in Fig. 1, a pilot-scale dynamic model of solvent-based PCC process is implemented in gCCS environment. Process model used in gCCS is based on the detailed dynamic model in Lawal et al [5]. The equipment parameters are kept the same at all the simulation scenarios. To validate the identified models in this manuscript, dynamic experimental data is not available in the current publications, the simulation data from gCCS was therefore used for model identification and comparison.

118 The working process of the considered solvent-based PCC plant is as follows: flue gas from a power plant or an industrial process is firstly cooled down to 40-50°C for a higher absorption performance, then it is fed into the bottom of absorber and comes 119 in contact with lean MEA solvent counter-currently. CO₂ is absorbed chemically and the treated gas leaves from the top side of 120 absorber. Rich (CO₂ concentration amine) solvent from the bottom of absorber is then pumped into the cross-heat exchanger and 121 preheated by hot lean (CO₂ concentration amine) solvent before entering the stripper. Low-pressure steam from power plants is 122 used in the reboiler. As a result of heat, the chemical bonds are thermally broken, releasing CO₂. The operational temperature of 123 the reboiler needs to be maintained within 383-393k to avoid amine degradation [21]. The vapor from the top of stripper is 124 125 condensed and separated in the condenser, water and amine are then refluxed back to stripper. Finally, the lean amine solution from the reboiler is cooled in the cross-heat exchanger by exchanging heat with rich CO₂ concentration amine before returning to 126 absorber. In general, a buffer tank containing a cooling coil is needed to keep water and MEA balance, as well as maintaining lean 127 solvent temperature. 128

- 129
- 130
- 131
- 132



Fig 1. Model topology in gCCS

The next step in this section is to select the manipulated and controlled variables. Lean solvent flowrate and steam flowrate are 136 the key variables influencing the characteristics of PCC plant. The control structures between lean solvent flowrate/steam flowrate 137 138 and capture rate/reboiler temperature are mostly discussed in previous studies [22, 23]. Moreover, this study considers the effects of condenser temperature and lean solvent temperature since they are closely related to the operation of PCC plant. Condenser 139 temperature is related to the purity of CO_2 product [24]. Therefore, for a higher concentration of CO_2 product, it is necessary to 140 141 maintain low condenser temperature since this has the benefit of reducing compression costs. Aroonwilas and Tontiwachwuthikul [25] reported that an increase in lean solvent temperature (at the Absorber top inlet) from 298K to 309K can lead to the increase 142 of CO₂ absorption ability. Beyond 309K, a further increase in the temperature to 318K may result in a reduction of overall mass 143 144 transfer coefficients. Therefore, lean solvent temperature is a vital parameter to be controlled for the best absorption performance. 145 For this reason, this paper mainly studies the dynamic characteristics of these key variables. As listed in Table 1, the lean solvent flow rate to absorber (u_1) , steam flowrate to reboiler (u_2) , cooling water flowrate to condenser (u_3) and cooling water flowrate to 146 147 cooler (u_4) are taken as 4 major manipulated variables, while capture rate (y_1), reboiler temperature (lean solvent side) (y_2), condenser temperature (y_3) and lean solvent temperature (y_4) are corresponding controlled variables. 148

| 1 | Δ | Q |
|---|----------|---|
| ┸ | – | 0 |

133 134

135

| Table 1. | Steady-state | controlled | values |
|----------|--------------|------------|--------|
|----------|--------------|------------|--------|

| | Manipulated variables | Controlled variables | Setpoint |
|----------------|-------------------------------------|--------------------------|----------|
| Control loop 1 | Lean solvent flowrate | Capture rate | / |
| Control loop 2 | Steam flowrate | Reboiler temperature | 383 K |
| Control loop 3 | Cooling water flowrate to condenser | Condenser temperature | 313.15 K |
| Control loop 4 | Cooling water flowrate to cooler | Lean solvent temperature | 313 K |

150 3.2. Steady-state values

151 This section carries out the steady-state analysis on these key variables. The steady-state values will provide preliminary 152 information on the steady-state features of PCC plant.

In this paper, capture rate is used as scheduling variable and to determine operating condition of PCC plant. This is due to its inherent nature of indicating the fulfillment of carbon absorption requirement in terms of environmental protection. Under this control circumstance (in Table 1), capture rate is varied in large scale while reboiler temperature, condenser temperature and lean solvent temperature are remained constant. Therefore, capture rate can be used to reveal the variation of working conditions for PCC plant and it is an important variable to be considered.

Steady-state simulations are carried out by adjusting capture rate setpoint from 50% to 95% in intervals of 5%. All the 158

manipulated and controlled variables are collected in Table 2 and plotted in Fig. 2. From the figure, it can be observed that all the 159

manipulated variables show an upward trend with increasing capture rate. Lean solvent flowrate and steam flowrate are 160 approximately proportional to the capture rate, while cooling water flowrate to cooler experiences a sharp increase when capture 161 rate reaches 90%. The cooling water flowrate to the condenser increases gradually until it reaches 90% capture rate. Above 90% 162 163 capture rate, the flowrate starts to decline.

| 1 | 64 |
|---|-------------------------|
| ㅗ | $\overline{\mathbf{U}}$ |

| Table 2. | Steady-state | manipulated value | ues |
|----------|--------------|-------------------|-----|
|----------|--------------|-------------------|-----|

| MVs | Lean solvent flow rate | Steam flowrate | Cooling water flowrate | Cooling water flowrate | |
|------------------|------------------------|----------------|------------------------|------------------------|--|
| | (kg/s) | (kg/s) | to condenser (kg/s) | to cooler (kg/s) | |
| 50% capture rate | 0.70801 | 0.0368226 | 0.162656 | 0.4388 | |
| 55% capture rate | 0.78350 | 0.0421757 | 0.166775 | 0.5239 | |
| 60% capture rate | 0.85924 | 0.0477303 | 0.170163 | 0.6234 | |
| 65% capture rate | 0.93545 | 0.0534870 | 0.172964 | 0.8099 | |
| 70% capture rate | 1.01332 | 0.0595083 | 0.175292 | 0.9964 | |
| 75% capture rate | 1.09216 | 0.0657443 | 0.177172 | 1.2504 | |
| 80% capture rate | 1.17361 | 0.0722941 | 0.178686 | 1.6191 | |
| 85% capture rate | 1.25718 | 0.0791476 | 0.179722 | 2.2229 | |
| 90% capture rate | 1.34627 | 0.0865317 | 0.180230 | 3.4151 | |
| 95% capture rate | 1.45123 | 0.0952342 | 0.179464 | 7.5704 | |



165 166

Fig. 2 Steady-state manipulated values

From Table 2 and Fig. 2, we can have a preliminary understanding on how these inputs will change at different capture rates. A 167 quantitative analysis of the process requires the identification of system models to provide dynamic parameter information, such 168 169 as time constant and settling time.

170 3.3. Local model identification

171 At different capture rates (50%, 60%, 70%, 80%, 90%, 95% capture rate), local models between manipulated variables and controlled variables (As listed in Tables 1) are obtained using System Identification Toolbox in MATLAB [14]. SISO transfer 172 function is selected as the model type because it is simple in identification and convenience in model analysis. 173

Such an identification technique utilizes input-output data to estimate mathematical model. The information provided by the 174 175 input-output data will influence the accuracy of the identified model. Considering the large time constant of PCC process, low 176 frequency pseudo-random binary sequence (PRBS) signal with a sampling time of 5 secs is designed as input to persistently excite the PCC system and provide enough information. An example of PRBS excitation data in lean solvent flowrate (u1) at 90% capture 177

178 rate is given in Fig. 3.

- To generate data for identification, the PCC process is in open loop. The input perturbation is applied to each input channel 179
- individually. All the identification and comparison data are generated in the gCCS modelling software. Before performing 180
- identification, all the data are pre-treated to remove mean value, outliers and noise. Singular value decomposition (SVD) on the 181
- Hankel matrix constructed from input-output data is conducted to estimate the model order [26]. A total number of 16 transfer 182
- functions (4 inputs by 4 outputs) are identified at every capture rate. The details of the identified model are available in the Appendix. 183





















Fig. 6. Comparison of cooler cooling water flowrate - reboiler temperature model at 90% capture rate

Step response tests are introduced to compare the identified local models with simulation data. As shown in Figs. 4-6, the local models are in satisfactory agreement with the simulation data from gCCS software. However, due to the space limits, only 3 models at 50%, 70% and 90% capture rate are chosen as examples. All the remaining models also give good results when compared with corresponding simulation data.

4. Open-loop step response analysis

Based on the identified local models in Section 3.3, open-loop step response tests are carried out under 50%, 60%, 70%, 80%, 90% and 95% capture rates for the major variables listed in Table 1. This study can provide the dynamic information of these variables, such as time constant and settling time. Besides, the step response test can inherently reveal how the input influences the corresponding output under varying operating conditions. This can enhance the understanding of nonlinear dynamic characteristics of PCC plant in a qualitative manner.

202 A relative variation of input Δu with a fixed value of 0.1 is performed in every scenario. The relative input Δu is expressed in 203 Equation (1).

204

$$\Delta u = \frac{u - \overline{u}}{\overline{u}} \tag{1}$$

where *u* denotes the absolute value introduced to input channel while \bar{u} denotes the steady-state values in different capture rates. Likewise, the model output is treated in the same way. The relative variations of outputs are depicted in Figs. 7-10.

All the step response curves are presented in the same benchmark in order to make reasonable performance comparison. To run the simulation, only one input is varied and the others remain constant.

209 4.1. Step changes in lean solvent flowrate

Using lean solvent flowrate to regulate capture rate is the most typical control option. It offers the advantage of faster response 210 and lower overshoot [27]. To gain insight into the transient behaviors of this loop, a relative step change Δu with a positive 211 amplitude of 0.1 is introduced to the lean solvent flowrate in the step time of 1000 secs. The relative output responses are displayed 212 213 in Fig. 7. It can be observed that capture rate increases sharply at the start of simulation for all cases, revealing the instant influence 214 of step change. Simultaneously, the solvent lean loading (mol CO₂ /mol MEA) also increases, leading to the decrease of capture rate after 1300 secs. Therefore, final value of the capture rate is lower than its initial value. This shows a typical non-minimum 215 phase feature, which may lead to the fluctuation of manipulated variable and it may also deteriorate control performance. Therefore, 216 an advanced control technique, e.g. pole assignment method, is advised to deal with this problem. Besides, the model time constants 217 at different capture rates are almost the same. Steady-state gains decrease with the increase of capture rate up to 90%, while the 218 219 gain in 95% capture rate has a sudden increase.



Fig. 7. Relative capture rate with the step change in lean solvent flowrate

4.2. Step changes in reboiler steam flowrate

Reboiler temperature is an important parameter which plays a key role in MEA regeneration process. It is regarded as the indicator of lean loading [23], which in turn reflects water makeup and capture rate controls. In this section, investigation of the effect of a positive increase in reboiler heat duty is carried out. The required reboiler heat duty is supplied from the low-pressure steam turbine in power plant. A relative increase of 0.1 in the steam flowrate is implemented in a relatively short period of time. Consequently, these is a significant increase in reboiler temperature, as observed in Fig. 8. The perturbations' amplitude raises gradually with increasing capture rate. It appears a relatively smooth change in the output response.

4.3. Step changes in cooling water flowrate to condenser

Condenser temperature is inversely related to the CO_2 purity [24]. Changing the flowrate of cooling water provides a potential option for the manipulation of the system. In this section, the open-loop performance of the condenser is investigated by increasing the flowrate of cooling water passing through the condenser. The process was simulated over a period of 50000 secs with a relative increase of 0.1 in the step time of 1000 secs. As shown in Fig. 9, condenser temperature reaches steady state after 40000 secs or 10 hrs, which indicates a very large inertia in the condenser. Furthermore, with decreasing capture rate, the settling time in condenser increases. Under this circumstance, condenser temperature will not be easily affected by other manipulated variables due to its large time constant.

4.4. Step changes in cooling water flowrate to cooler

According to [25], lean solvent temperature will affect the overall mass transfer coefficients of absorber column. Therefore, lean 238 solvent temperature needs to be controlled for the higher absorption performance. In this paper, we used a counter-current heat 239 exchanger (As shown in Fig. 1) to maintain lean solvent temperature. This may reduce the system ability to resist disturbances in 240 lean solvent flowrate, but the time necessary for controlling lean solvent temperature can be reduced, since it doesn't have massive 241 liquid storage. Fig. 10 shows the output response in the presence of step change in the flowrate of cooling water entering the cooler. 242 The amplitude of a relative step change in flowrate of cooling water is 0.1. It can be observed that the cooling water flowrate has 243 an instant influence on the lean solvent temperature. This is due to the large heat transfer coefficient and heat transfer area in the 244 cooler. After that, the lean solvent temperature decreases gradually, this is influenced by the decrease in reboiler temperature. On 245 the other hand, the model in 50% capture rate has the highest steady state gains and longest settling time. With the increase of 246 247 capture rate, these two parameters decrease dramatically. Preliminary results can be obtained to indicate a much stronger 248 nonlinearity in the cooler compared with other mentioned units.

249

220 221











Fig 10. Relative lean solvent temperature with the step change in cooling water flowrate to cooler

258 4.5. Sensitivity analysis

A sensitivity analysis is carried out using the input variable perturbation method. This analysis provides a quantitative evaluation 259 of outputs relating to possible changes in input variables. The relative input data along with their sensitivity index (relative outputs) 260 261 are portrayed in Table 3. Sensitivity analysis can provide information in the process dynamics and are able to calculate process gains. According to the results, there is an inverse correlation between lean solvent flowrate (u_1) and capture rate (y_1) . A similar 262 correlation can be found between condenser cooling water flowrate (u_3) and condenser temperature (y_3) as well as cooler cooling 263 water flowrate (u₄) and lean solvent temperature (y₄). u₁ has a strong effect to its corresponding output y₁, while u₂, u₃, u₄ are much 264 less influential towards their outputs (y_2 , y_3 , y_4). This indicates that the controller gain designed for u_1 - v_1 model should be smaller 265 than those in u₂-y₂, u₃-y₃ and u₄-y₄ models. Furthermore, the sensitivity index changes with the variation of capture rates. Indexes 266 267 of models in u₁-y₁, u₂-y₂ and u₃-y₃ loops has a narrow change at different capture rates, while the index in u₄-y₄ loop varies rapidly. This supports the nonlinearity analysis obtained in Section 5.1. 268

269

Table 3. Sensitivity analysis of 4 SISO models

| In most of () | Stan alamaaa | Sensitivity Index (Output Δy_i) | | | | | | |
|--------------------------------------|--------------|------------------------------------------|----------------------|--------------------|----------------------|----------------------|--------------------|-----------------|
| Inputs(<i>u</i> _i) Step | Step changes | $\Delta y_{i}(50\%)$ | $\Delta y_{i}(60\%)$ | $\Delta y_i(70\%)$ | $\Delta y_{i}(80\%)$ | $\Delta y_{i}(90\%)$ | Δy_i (95%) | |
| $\triangle u_1$ | 10% | -0.1729 | -0.1966 | -0.2178 | -0.2355 | -0.2447 | -0.2360 | $\triangle y_1$ |
| $\triangle u_2$ | 10% | 0.0077 | 0.0086 | 0.0097 | 0.0106 | 0.0123 | 0.0147 | $\triangle y_2$ |
| $\triangle u_3$ | 10% | -0.0359 | -0.0321 | -0.0291 | -0.0261 | -0.0231 | -0.0219 | $\triangle y_3$ |
| $\triangle u_4$ | 10% | -0.0065 | -0.0054 | 0.0039 | -0.0026 | -0.0013 | -0.00063 | $\triangle y_4$ |

270 **5. Nonlinearity analysis**

With variation in operating conditions, dynamic characteristics of solvent-based PCC plant may change and exhibit an inherent nonlinearity. Using the local models developed in Section 3.3, this section provides a nonlinearity analysis based on gap metric to quantify the nonlinearity degree of PCC process (as modelled in gCCS). Compared with [10], this paper put more key variables (As listed in Table 1) in consideration to investigate their nonlinear characteristics in a quantitative manner. Nonlinearity measurement is also carried out in SISO model to reveal the relationship between input and output variables at varying capture rates. These discussions set this section apart from the similar work in [10].

The notion of gap to measure distance between nonlinear system was explained in Zams and El-Sakkary [28]. In the same paper, gap metric was firstly introduced to capture the uncertainty in feedback system. Later it was found that gap metric is more suitable to measure the distance between two linear systems than using a norm-based metric calculation [29]. This section is to use a differential gap metric defined in [30] to measure the distance between two linear models. The method used in [30] is more applicable and feasible in real process. The gap metric is defined by Equation (2):

$$\delta_{d}(N_{1}, N_{2}) = \max\left\{\vec{\delta}_{d}(N_{1}, N_{2}), \vec{\delta}_{d}(N_{2}, N_{1})\right\}$$
(2)

283 where:

282

284
$$\vec{\delta}_{d}(N_{1}, N_{2}) = \sup_{r_{1}} \inf_{r_{2}} \delta_{d}(L_{r_{1}}N_{1}, L_{r_{2}}N_{2})$$

285
$$\delta_{d}(L_{f_{1}}N_{1},L_{f_{2}}N_{2}) = \left\| \Pi_{G(L_{f_{1}}N_{1})} - \Pi_{G(L_{f_{2}}N_{2})} \right\|$$

286 $L_{r_i}N_i$ represents the linear approximation model of N_i at the point of r_i and L denotes linearization. Π represents the 287 rectangular projection and G is the subspace of the product Hilbert space. SISO transfer functions will be used in Section 5.1 and 288 MIMO transfer function matrix will be used in Section 5.2.

- 289
- 290

Due to space limits, only the nonlinear evaluation of models in 4 major control loops (as listed in Table 1) is presented in this section. From the results displayed in Fig. 11, it is clear that nonlinearity degrees for lean solvent flowrate (u_1) - capture rate (y_1) , steam flowrate (u_2) – reboiler temperature (y_2) , condenser cooling water flowrate (u_3) – condenser temperature (y_3) models are very small, while the degree for cooler cooling water flowrate (u_4) – lean solvent flowrate (y_4) model is very large compared with the other 3 models.

This demonstrates that the nonlinearities of models in u_1 - y_1 , u_2 - y_2 , u_3 - y_3 control loops are weak and evenly distributed within the 50%-95% capture rate operating range. The models in these 3 loops are close to the models in their adjacent working condition. However, the disparity in model output results (with same inputs) between low and high working conditions (e.g. 50% capture rate and 90% capture rate) can be very large. The u_4 - y_4 model shows a strong nonlinear behavior with varying working conditions. The results obtained in this section are in consistent with the open-loop step response tests in Figs. 7-10.

302



303 304

309

Fig 11. Nonlinearity degree of SISO system

305 5.2. Nonlinearity analysis of MIMO system

306 In this section, a nonlinear gap measurement of MIMO system is attempted to discover the nonlinear characteristics of the overall 307 system. The 4-input-4-output system is expressed in Equation (3), where g_{ij} denotes the transfer function model in the loop from 308 u_j to y_i . Details of transfer function g_{ij} can be found in the Appendix.

310 System 1:
$$\begin{bmatrix} y_1 \\ y_2 \\ y_3 \\ y_4 \end{bmatrix} = \begin{bmatrix} g_{11} & g_{12} & g_{13} & g_{14} \\ g_{21} & g_{22} & g_{23} & g_{24} \\ g_{31} & g_{32} & g_{33} & g_{34} \\ g_{41} & g_{42} & g_{43} & g_{44} \end{bmatrix} \begin{bmatrix} u_1 \\ u_2 \\ u_3 \\ u_4 \end{bmatrix}$$
(3)

The results of nonlinear evaluations of System 1 are portrayed in Fig. 12. From this figure, the nonlinearity degree is very large in any two adjacent load conditions. This may be due to the strong nonlinear properties in the model of u_4 - y_4 loop. This reveals that the controller tuned for a certain capture rate (e.g. 90% capture rate) can only function in the vicinity of the mentioned capture rate. The control performance may deteriorate if this tuned controller is used for much lower capture rates scenario.

According to the open-loop step response test and nonlinearity analysis, PCC plant exhibits strong nonlinearity at varying capture rates. This indicates that the linear local models are not sufficiently enough to simulate the nonlinear dynamic characteristics. Given this context, it is necessary to develop a piece-wise model by a combination of linear local model in order to predict the nonlinear features of PCC process. Using more local models to develop such a piece-wise model will give more accurate prediction results

- 319 [19]. However, this will also increase the complexity in model implementation and calculation. Considering its evenly distributed
- nonlinearity of MIMO system, the local models in 50%, 60%, 70%, 80%, 90% and 95% capture rate are therefore selected to form
- 321 the piece-wise model.





324 6. Piece-wise model and model comparison

325 6.1. Fuzzy-based piece-wise model

According to the open-loop step response test and nonlinearity analysis of PCC process in the previous sections, the model dynamic characteristics differ significantly with varying capture rates. To this end, the identified models in Section 3.3 are selected to develop the piece-wise model in order to predict the nonlinear characteristics of PCC process. The unification of the local linear models is based on the concept of fuzzy sets theory [31]. This makes the fuzzy model simpler in application.

In this section, the Takagi and Sugeno (TS) fuzzy modelling method [32] is adopted to combine the local linear models. The first step is to determine the fuzzy variables and their working range. Capture rate is used as scheduling variable. Owing to the targets to carry out in this study, capture rate is studied from 50% to 95%. In Fig. 13, a six-rule fuzzy triangular membership function of lean solvent flowrate – capture rate model is given as an example.



334 335

322 323

336

Fig. 13. Membership functions of lean solvent flowrate - capture rate model

337 With the membership function, the output of piece-wise model can be derived from Equation (4):

338
$$y_{Model} = h_1(mf_{50}) \times y_{50} + h_2(mf_{60}) \times y_{60} + h_3(mf_{70}) \times y_{70} + h_4(mf_{80}) \times y_{80} + h_5(mf_{90}) \times y_{90} + h_6(mf_{95}) \times y_{95}$$
(4)

where $h_i(mf_j)$ denotes the triangular weighting functions and y_j denotes the output of local transfer function in the *j* th capture rate. $h_i(mf_j)$ can be obtained from the triangular functions in Figure. 13 and $\Sigma h_i(mf_j)$ equals to 1.

341 Simulink was used to develop the fuzzy-based piece-wise model. The details of the model structures are described in Fig. 14.
342 The models of other control loops, e.g. steam flowrate to reboiler temperature model, condenser cooling water flowrate to
343 condenser temperature model and cooler cooling water flowrate to lean solvent temperature model, can be obtained in the same
344 manner.



353

354

355

356 357

358

359

360

Fig. 14. The piece-wise lean solvent flowrate - capture rate model in Simulink workspace

347 6.2. Model comparison

348 In this section, the proposed piece-wise models are compared with local linear models and simulation data in the wide-range 349 variation of capture rates. The significance of this comparison is to demonstrate the accuracy of developed piece-wise model in 350 predicting performance of solvent-based PCC plant at different operating conditions.

Due to space limits, the piece-wise models in main 4 control loops (in Table 1) are presented as examples. For simplicity, 4 scenarios are presented in this section, which include:

- The piece-wise lean solvent flowrate to capture rate model is compared with local model (developed at 90% capture rate) and simulation data (collected around 92.5% capture rate).
- The piece-wise steam flowrate to reboiler temperature model is compared with local model (developed at 70% capture rate) and simulation data (collected around 75% capture rate).
- The piece-wise condenser cooling water flowrate to condenser temperature model is compared with local model (developed at 60% capture rate) and simulation data (collected around 65% capture rate).
 - The piece-wise cooler cooling water flowrate to lean solvent temperature model is compared with local model (developed at 50% capture rate) and simulation data (collected around 55% capture rate).

Local models were identified for a given set of capture rates (in Section 3.3) and then combined to obtain the piece-wise models (in Section 6.1). The piece-wise models are compared with simulation data from other capture rates different from the given set. Therefore, the validity of the piece-wise model in simulating the performance of PCC plant at varying operating conditions is proven.

The comparison results are given in Figs. 15-18. As shown in Fig. 15 and Fig. 18, it is clear that the piece-wise models are in good agreement with simulation data and are more accurate compared to local models. The results indicate a high level of divergence between piecewise and the local models. However, the disparity between piecewise and the local model in Fig. 15 is much narrow in comparison to the result in Fig. 18. This reveals a much stronger nonlinearity in cooler. A local linear model is not enough to simulate the nonlinear features of cooler unit.

```
Nevertheless, from Fig. 16-17, it is obvious that the output results of the local models are close to those of the piece-wise models.
This is due to the weak nonlinear effects of reboiler and condenser on the process of the PCC plant. The weakness of their nonlinear
effects can be demonstrated by their large time constant, which slows down the variation of dynamic feature with the changing of
input variables. This means that the number of local linear models for piece-wise combination can be reduced.
```

In conclusion, the proposed fuzzy-based piece-wise model can satisfactorily simulate the dynamic characteristics of PCC plant over a wide range of operating conditions. The comparisons in these figures reveal strong nonlinearity in cooler, while much weaker nonlinearities in reboiler and condenser. These results are in in consistent with the conclusion displayed in Fig. 11.



Fig. 15. Comparison of piece-wise lean solvent flowrate - capture rate model around 92.5% capture rate



Fig. 16. Comparison of piece-wise steam flowrate - reboiler temperature model around 75% capture rate



Fig. 17. Comparison of piece-wise condenser cooling water flowrate - condenser temperature model around 65% capture rate



Fig. 18. Comparison of piece-wise cooler cooling water flowrate - lean solvent temperature model around 55% capture rate

389 **7. Conclusion**

On the basis of system identification and nonlinearity analysis, this paper developed a piece-wise model to simulate the nonlinear 390 dynamic characteristics of the solvent-based PCC process. The piece-wise model shows satisfactory agreement with comparison 391 392 data and it is more accurate compared with local models. Using simulation data from gCCS software, SISO local transfer function models were firstly identified at every capture rate scenario. Open-loop step response tests were then introduced to show the 393 394 dynamic features of PCC plant under changing operating conditions. Lean solvent flowrate was found to be very influential on 395 capture rate. The nonlinearity analysis was then carried out using differential gap metric. It was found that the nonlinearity degrees in lean solvent flowrate - capture rate model, steam flowrate - reboiler temperature model and condenser cooling water - condenser 396 temperature model are very small for any two adjacent operating conditions. However, the cooler cooling water flowrate to lean 397 solvent temperature model produces results which exhibit high divergence. Nonlinearity degrees of MIMO PCC system are proven 398 399 to be equally high in any two operating conditions. The results of this study will provide an enhanced knowledge of transient performance of PCC process and provide guidance for the flexible controller design. 400

401 Acknowledgements

The authors would like to acknowledge the National Natural Science Foundation of China (NSFC) under Grant 51506029, the Natural Science Foundation of Jiangsu Province, China under Grant BK20150631, China Postdoctoral Science Foundation, EU FP7 International Staff Research Exchange Scheme on power plant and carbon capture (Ref: PIRSES-GA-2013-612230) and China scholarship council (CSC no:201606090176) for funding this work.

406 Appendix. Supplementary models

407 Supplementary models of solvent-based PCC process from 50% capture rate to 95% capture rate in each SISO loop are available.

408 References

- 409 [1] Coninck HC, Loos MA, Metz B, Davidson O, Meyer LA. IPCC special report on carbon dioxide capture and storage.
 410 Intergovernmental Panel on Climate Change 2005.
- 411 [2] DTI UK. Meeting the energy challenge: A white paper on energy. Department for Trade and Industry, Cm7124, 2007.
- Wang M, Lawal A, Stephenson P, Sidders J, Ramshaw C. Post-combustion CO₂ capture with chemical absorption: a state-of the-art review. Chemical Engineering Research and Design 2011; 89: 1609-1624.
- 414 [4] Abu-Zahra MR, Schneiders LH, Niederer JP, Feron PH, Versteeg GF. CO₂ capture from power plants: Part II. A parametric

- study of the economic performance based on mono-ethanolamine. International Journal of Greenhouse Gas Control 2007;
 1(2): 135-142.
- Lawal A, Wang M, Stephenson P, Koumpouras G, Yeung H. Dynamic modelling and analysis of post-combustion CO₂
 chemical absorption process for coal-fired power plants. Fuel 2010; 89: 2791-2801.
- [6] Lawal A, Wang M, Stephenson P, Yeung H. Dynamic modelling of CO₂ absorption for post combustion capture in coal-fired
 power plants. Fuel 2009; 88: 2455-2462.
- [7] Lawal A, Wang M, Stephenson P, Obi O. Demonstrating full-scale post-combustion CO₂ capture for coal-fired power plants
 through dynamic modelling and simulation. Fuel 2012; 101: 115-128.
- 423 [8] Dugas, ER. Pilot plant study of carbon dioxide capture by aqueous mono-ethanolamine. M.S.E. Thesis. University of Texas,
 424 Austin, USA; 2006
- [9] Biliyok C, Lawal A, Wang M, Seibert F. Dynamic modelling, validation and analysis of post-combustion chemical absorption
 CO2 capture plant. International Journal of Greenhouse Gas Control 2012; 9: 428-445.
- [10] Wu X, Shen J, Li Y, Wang M, Lawal A. Flexible operation of post-combustion solvent-based carbon capture for coal-fired
 power plants using multi-model predictive control: A simulation study. Fuel 2018; 220: 931-941.
- [11] Kvamsdal HM, Jakobsen JP, Hoff KA. Dynamic modeling and simulation of a CO₂ absorber column for post-combustion
 CO₂ capture. Chemical Engineering and Processing: Process Intensification 2009; 48(1): 135-144.
- [12] Ziaii S, Rochelle GT, Edgar TF. Dynamic modeling to minimize energy use for CO₂ capture in power plants by aqueous
 mono-ethanolamine. Industrial & Engineering Chemistry Research 2009; 48(13): 6105-6111.
- [13] Arce A, Mac Dowell N, Shah N Vega LF. Flexible operation of solvent regeneration systems for CO₂ capture processes using
 advanced control techniques: Towards operational cost minimization. International Journal of Greenhouse Gas Control 2012;
 11: 236-250.
- 436 [14] L. Ljung. System Identification Toolbox. For use with Matlab 1995.
- [15] Manaf NA, Cousins A, Feron P, Abbas A. Dynamic modelling, identification and preliminary control analysis of an amine based post-combustion CO2 capture pilot plant. Journal of Cleaner Production 2016; 113: 635-653.
- [16] Mac Dowell N, Samsatli NJ, Shah N. Dynamic modelling and analysis of an amine-based post-combustion CO₂ capture
 absorption column. International Journal of Greenhouse Gas Control 2013; 12: 247-258.
- [17] Li F, Zhang J, Oko E, Wang M. Modelling of a post-combustion CO2 capture process using neural networks. Fuel 2015; 151:
 156-163.
- [18] Hamming R. Numerical methods for scientists and engineers. Courier Corporation 2012.
- [19] Sontag E. Nonlinear regulation: The piece-wise linear approach. IEEE Transactions on automatic control 1981; 26(2): 346358.
- 446 [20] Process Systems Enterprise UK. gCCS overview, http://www.psenterprise.com/ products/gccs.html
- 447 [21] Harun N. Dynamic simulation of MEA absorption process for CO₂ capture from power plant. PhD thesis, University of
 448 Waterloo; 2012.
- [22] Lin YJ, Chang CC, Wong DSH, Jang SS, Ou JJ. Control strategies for flexible operation of power plant with CO₂ capture
 plant. AIChE Journal 2012; 58: 2697-2704.
- [23] Lin YJ, Pan TH, Wong DSH, Jang SS, Chi YW, Yeh CH. Plantwide control of CO₂ capture by absorption and stripping using
 mono-ethanolamine solution. Industrial & Engineering Chemistry Research 2010; 50: 1338-1345.
- [24] Nittaya T, Douglas PL, Croiset E, Luis A, Sandoval R. Dynamic modelling and control of MEA absorption processes for CO₂
 capture from power plants. Fuel 2014; 116: 672-691.
- [25] Aroonwilas A, Tontiwachwuthikul P, Chakma A. Effects of operating and design parameters on CO₂ absorption in columns
 with structured packings. Separation and Purification Technology 2001; 24(3): 403-411.
- 457 [26] Yin H, Zhu Z, Ding F. Model order determination using the Hankel matrix of impulse responses. Applied Mathematics
 458 Letters 2011; 24(5): 797-802.
- [27] Mechleri E, Lawal A, Ramos A, Davison J, Mac Dowell N. Process control strategies for flexible operation of post combustion CO₂ capture plants. International Journal of Greenhouse Gas Control 2017; 57: 14-25.
- 461 [28] El-Sakkary A. The gap metric: Robustness of stabilization of feedback systems. IEEE Transactions on Automatic Control

- 462 1985; 30(3): 240-247.
- 463 [29] Georgiou TT, Smith MC. Optimal robustness in the gap metric. IEEE Transactions on Automatic Control 1990; 36(6): 673-
- 464 686.
- [30] Georgiou TT. Differential stability and robust control of nonlinear systems. Signals and Systems 1993; 6(4): 289-306.
- 466 [31] Zimmermann HJ. Fuzzy set theory and its applications. Springer Science & Business Media, 2011.
- 467 [32] Takagi T, Sugeno M. Fuzzy identification of systems and its applications to modeling and control. IEEE Transactions on
 468 Systems, Man and Cybernetics 1985; 1: 116-132.
- 469
- 470
- 471