Dynamic Modeling of MEA-based CO2 Capture in Biomass-fired CHP Plants

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Abstract

To achieve the targets set by the Paris Agreement, CO₂ emissions reduction alone is no longer sufficient, and carbon dioxide removal is required by an extensive deployment of negative emission technologies (NETs). Bioenergy with CO₂ capture and storage (BECCS) is one of the most promising NETs. Integrating BECCS with biomass-fired and waste-fired combined heat and power (bio-CHP and w-CHP) plants is considered the most feasible solution. However, bio/w-CHP plants are characterized by high fluctuations in operation, which can result in more dynamic variations of flue gas flowrates and compositions and available heat for CO₂ capture. Such changes can clearly affect the performance of CO₂ capture; therefore, doing dynamic simulations for CO₂ capture becomes crucial. This thesis aims to investigate the performance of different dynamic physical model-based approaches, identify their quantitative differences and provide suggestions for approach selection. Three physical model-based approaches include the ideal static approach, the dynamic approach without control, and the dynamic approach with control. The operating data from an actual waste CHP plant is employed and various cases are defined considering different critical operating parameters of bio/w-CHP plants. Although apparent differences can be observed in the results from different approaches, the differences become smaller when there are both increases and decreases in the variations of parameters. The results will provide valuable recommendations about selecting the dynamic modeling approach through the whole chain of the design, operation, and optimization of CO₂ capture. In addition, this thesis also tests the data-driven modeling approach, Informer, and results show Informer can accurately predict the CO₂ capture rate and energy penalty of dynamic CO₂ capture.
To my family
I am deeply grateful that I can work on this project. My deep and sincere gratitude goes to all my supervisors, Prof. Hailong Li, Dr. Jan Skvaril, and Prof. Eva Thorin, for their guidance and support. Over the last two years, they have helped me formulate and improve my research and shown continuous suggestions and patience in reviewing my papers. I would like to thank Dr. Lara Carvalho for reviewing my licentiate thesis. I also want to thank the good colleagues and friends I met at Mälardalens University. Thanks a lot for all your support in both research and life!

I gratefully acknowledge the Swedish Energy Agency (Energimyndigheten) for the financial support. I am also grateful for the support from the collaboration partners of Stockholm Exergi, Mälarenergi, and Eskilstuna Strängnäs Energi och Miljö. The discussions with them can always enlighten me and improve my research.
Summary

Global warming is a significant threat to our planet. Adopting the Paris Agreement is a global action that aims to reduce greenhouse gas (GHG) emissions. An extensive deployment of negative emission technologies (NETs) is required to achieve the targets set by the Paris Agreement. Bioenergy with carbon capture and storage (BECCS) is emerging as one of the most promising NETs. Among different biomass utilization processes, integrating BECCS with biomass-fired and waste-fired combined heat and power (bio-CHP and w-CHP) plants has been considered the most feasible solution. Bio/w-CHP plants are characterized by high fluctuations in operation, which can result in more dynamic variations of flue gas (FG) flowrates and compositions and available heat for CO₂ capture. Such changes can clearly affect the performance of CO₂ capture; therefore, doing dynamic simulations becomes crucial.

This thesis aims to investigate the performance of different dynamic physical model-based approaches and provide suggestions for approach selection. In addition, the data-driven modeling approach, which is an emerging technology, has also been tested.

Three physical model-based approaches include the ideal static model (IST), the dynamic approach without control (Dw/oC), and the dynamic approach with control (DwC). To compare their performance, the operating data from an actual waste CHP plant is employed. Various cases have been defined considering different critical operating parameters, including the FG flowrate, the CO₂ concentration (CO₂vol%), and the available heat for CO₂ capture. Apparent differences can be observed in the results from different approaches. For example, when the CO₂vol% drops from 15.7 % to 9.7 % (about 38 %) within 4 hours, the difference in the captured CO₂ can be up to 22% between DwC and Dw/oC. It is worth noting that when there are both increases and decreases in the variations of parameters, the differences become smaller.

Based on the comparison, the recommendations on approaches have been summarized. Dw/oC is recommended for checking the boundary of safety operation by the response analysis. DwC is recommended for designing the control system, observing the flexible dynamic operation, estimating the short-term CO₂ capture potential, and optimizing the hourly dynamic operation. IST is recommended for estimating the long-term CO₂ capture potential, and optimizing the long-term dynamic operation when the input parameters vary not as often as hourly.
A data-driven model, Informer, is developed to model CO₂ capture dynamically. The dataset is generated by using a physical model. The FG flowrate, the CO₂ vol%, the lean solvent flowrate, and the available heat for CO₂ capture are employed as input parameters, and the CO₂ capture rate and the energy penalty are chosen as outputs. The results show that Informer can accurately predict dynamic CO₂ capture. The mean absolute percentage error (MAPE) was found to be 6.2% and 2.7% for predicting the CO₂ capture rate and energy penalty, respectively.
Swedish Summary


Denna avhandling syftar till att undersöka hur olika dynamiska fysiska modellbaserade tillvägagångssätt fungerar och ge förslag på val av tillvägagångssätt. Dessutom har den datadrivna modelleringen, som är en framväxande teknologi, också testats.

Tre fysiska modellbaserade tillvägagångssätt inkluderar den ideala statiska modellen (IST), den dynamiska metoden utan kontroll (Dw/oC) och den dynamiska metoden med kontroll (DwC). För att jämföra deras prestanda används driftsdata från ett verkligt avfallskraftvärmeverk. Olika fall har definierats med hänsyn till olika kritiska driftsparametrar, inklusive FG-flödet, CO2-koncentrationen (CO2vol%) och tillgänglig värme för CO2-avskiljning. Synbara skillnader kan observeras i resultaten från olika tillvägagångssätt. Till exempel, när CO2vol% sjunker från 15.7% till 9.7% (cirka 38%) inom 4 timmar, kan skillnaden i fångad CO2 vara upp till 22% mellan DwC och Dw/oC. Det är värt att notera att när det finns både ökningar och minskningar av variationerna av parametrar, blir skillnaderna mindre.

Baserat på jämförelsen har rekommendationerna om tillvägagångssätt sammanfattats. Dw/oC rekommenderas för att kontrollera gränsen för säkerhetsdrift genom svarsanalysen. DwC rekommenderas för att utforma styrsystemet, observera den flexibla dynamiska driften, uppskatta den kortsiktiga CO2-avskiljningspotentialen och optimera den dynamiska driften per timme. IST rekommenderas för att uppskatta den långsiktiga CO2-avskiljningspotentialen...
och för att optimera den långsiktiga dynamiska driften när ingångsparamet-
rarna inte varierar så ofta som varje timme.

En datadriven modell, Informer, är utvecklad för att modellera CO₂-fångst
dynamiskt. Datauppsättningen genereras med hjälp av en fysisk modell. FG-
flödet, CO₂-vol%, det magra lösningsmedelsflödet och tillgänglig värme för
CO₂-avskiljning används som ingångsparametrar, och CO₂-infångning hastig-
heten och energi straffet väljs som utdata. Resultaten visar att Informer exakt
can förutsäga dynamisk CO₂-avskiljning. Det genomsnittliga absoluta procentuella felet (MAPE) visade sig vara 6.2 % och 2.7 % för att förutsäga CO₂-
infångning hastigheten respektive energi straffet.
List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.


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Contents

Acknowledgements...........................................................................................................i
Summary ............................................................................................................................. iii
Swedish Summary .............................................................................................................. v
List of Papers .................................................................................................................... vii
Contents ............................................................................................................................ ix
List of Figures ................................................................................................................... xi
List of Tables .................................................................................................................... xii
Nomenclature ................................................................................................................... xiii

1  Introduction .................................................................................................................. 1
  1.1  Background ............................................................................................................. 1
  1.2  Knowledge gaps and challenges ......................................................................... 2
  1.3  Scope and objectives ......................................................................................... 3
  1.4  Methodology ....................................................................................................... 3
  1.5  Thesis structure ................................................................................................. 4

2  Literature review ......................................................................................................... 7
  2.1  Integrating BECCS in biomass/waste CHP plants ............................................ 7
  2.2  Physical model-based approaches to CO₂ capture ........................................ 8
  2.3  Data-driven approaches to CO₂ capture ......................................................... 10

3  Approaches to modeling dynamic CO₂ capture in bio/w-CHP plants. 11
  3.1  Physical model-based approaches to CO₂ capture ......................................... 11
      3.1.1  Approach of ideal static model (IST) ..................................................... 11
      3.1.2  Dynamic approach without control (Dw/oC) ....................................... 12
      3.1.3  Dynamic approach with control (Dw/C) ............................................. 13
  3.2  Case study ........................................................................................................... 15
      3.2.1  Data from the CHP plant and model scale-up ..................................... 15
      3.2.2  Critical operating parameters ......................................................... 17
      3.2.3  Key performance indicators ............................................................. 20
  3.3  The data-driven approach ............................................................................... 20
      3.3.1  Dataset preparation ............................................................................ 21

3.3.2 The architecture of Informer.............................................................. 22

4 Results and discussions ................................................................. 25
  4.1 Comparison on CO₂ capture performance.................................. 25
  4.1.1 Case 1: Variation of the FG flowrate................................. 25
  4.1.2 Case 2: Variation of the CO₂vol%...................................... 27
  4.1.3 Case 3: Variation of both FG flowrate and CO₂vol%......... 28
  4.1.4 Case 4: Variation of the available heat............................ 29
  4.1.5 Case 5: Variation of the FG and available heat................. 30
  4.1.6 Comparison on the overall performance.......................... 31
  4.1.7 Influence of time step (TS).................................................. 32
  4.2 Selection of modeling approaches....................................... 34
  4.3 Performance of Informer model........................................... 36

5 Summary of papers ................................................................. 39
  5.1 Paper I.................................................................................. 39
  5.1.1 My contribution............................................................... 39
  5.1.2 Results and discussions.................................................. 39
  5.2 Paper II............................................................................... 40
  5.2.1 My contribution............................................................... 40
  5.2.2 Results and discussions.................................................. 40

6 Conclusions ............................................................................... 41

7 Future work............................................................................... 43

References...................................................................................... 45

Appendix......................................................................................... 49
  A1: Validation of the steady state model in Aspen plus .......... 49
  A2: Validation of the dynamic model in Aspen HYSYS.......... 50
  A3: Methods of the model scale-up........................................ 52
List of Figures

Figure 1.1: The methodological approach of the thesis .................................. 4
Figure 3.1: Rate-based simulation of MEA-based CO$_2$ capture in Aspen Plus. ................................................................. 12
Figure 3.2: MEA-based CO$_2$ capture without control strategy in Aspen HYSYS ................................................................................................. 13
Figure 3.3: MEA-based CO$_2$ capture with control strategy .................... 14
Figure 3.4: FG data from a w-CHP plant .................................................. 16
Figure 3.5: Variation of FG flowrate in Case 1 ........................................... 17
Figure 3.6: Variation of CO$_2$vol% in Case 2 ............................................. 18
Figure 3.7: Variation of FG flowrate and CO$_2$vol% in Case 3 ............... 18
Figure 3.8: Variation of the available heat in Case 4 ............................... 18
Figure 3.9: Variation of the available heat in Case 5 ............................... 18
Figure 3.10: Input and output parameters of the generated dataset ........ 21
Figure 3.11: Excitation signal of the four input parameters (a) Reboiler duty (b) Lean MEA solution flowrate (c) FG CO$_2$vol% (d) FG flowrate. ... 22
Figure 4.1: Model comparison under the variations of FG flowrate ......... 26
Figure 4.2: Model comparison under the variations of FG CO$_2$vol% ....... 28
Figure 4.3: Model comparison under variations of both FG flowrate and CO$_2$vol%. ................................................................. 29
Figure 4.4: Model comparison under the variations of available heat ....... 30
Figure 4.5: Model comparison under the variations of FG and available heat. ................................................................................. 31
Figure 4.6: Influence of time step on dynamic changes of KPIs in Case 5-1. ........................................................................................................ 33
Figure 4.7: Influence of time step on the accumulated amount of captured CO$_2$. .............................................................. 34
Figure 4.8: FG flowrate and CO$_2$vol% of DwC and IST of one day .......... 35
Figure 4.9: The difference in captured CO$_2$ between DwC and IST ......... 36
Figure 4.10: The result of CO$_2$ capture rate ........................................... 37
Figure 4.11: The result of the energy penalty ........................................... 37
List of Tables

Table 2.1: Three approaches to dynamic modeling of CO₂ capture. .............. 8
Table 3.1: Summary of model parameters and column settings (K. Li et al., 2015). ......................................................................................................................... 12
Table 3.2: Dynamic model parameters (Harun, 2012). ........................................ 13
Table 3.3: Manipulated and controlled variables and target of controllers (Harun et al., 2012; Nittaya et al., 2014). ................................................................. 14
Table 3.4: The scaled-up capture system and other input parameters for the case study. ............................................................................................................. 16
Table 3.5: An overview of the defined cases. ......................................................... 19
Table 4.1: Comparison of different approaches for overall performance. .... 32
Nomenclature

Abbreviations

BA-NN  Bootstrap aggregated neural networks
BECCS  Bioenergy with carbon capture and storage
Bio-CHP Biomass fired combined heat and power
CDR  Carbon dioxide removal
CHP  Combined heat and power
CO2vol%  CO2 volume concentration
COP 28  The 28th Conference of the Parties
DH  District heating
DwC  Dynamic model with control
Dw/oC  Dynamic model without control
FG  Flue gas
FIC  Flowrate integral controllers
GHGs Greenhouse gases
IST  Ideal static model
KPIs Key performance indicators
LIC  Liquid level integral controllers
L/G ratio Liquid-to-gas ratio
LTSF  Long-term time series forecasting
MAPE Mean absolute percentage error
MEA Monoethanolamine
MEA-CA MEA-based chemical absorption
ML  Machine learning
MPC  Model predictive control
MSE Mean squared error
NETs Negative emissions technologies
NRDC Natural Resources Defence Council
PCC Post-combustion capture
PI Proportional–integral
PPA Power purchase agreement
TIC Temperature integral controllers
TOU Time of use
TS Time step
W-CHP Waste fired combined heat and power
Symbols

\( CO_{2pro} \)  Amount of CO\(_2\) in Product
\( CO_{2FG} \)  Amount of CO\(_2\) in Flue gas
\( i \)  The \( i^{th} \) sample
\( n \)  Sample size
\( Q_{reb} \)  Reboiler duty
\( t \)  Integral time
\( T \)  Temperature
\( u_1 \)  FG flow rate
\( u_2 \)  FG CO\(_2\) concentration
\( u_3 \)  Lean solution flowrate
\( u_4 \)  Reboiler duty
\( y_1 \)  CO\(_2\) capture rate
\( y_2 \)  Energy penalty per unit captured CO\(_2\)
\( \hat{y} \)  Actual value
\( y \)  Predicted value
1 Introduction

1.1 Background

For the last 40 years, the Earth’s global average temperature has been increasing fast, with more than 0.2 °C per decade. Emissions of greenhouse gases (GHGs) from fossil fuel burning are mostly blamed for global warming, and CO$_2$ is the most important GHG. Based on the Natural Resources Defence Council (NRDC), global warming could threaten climate systems by exacerbating drought, floods, and wildfires, jeopardizing our air, water, and food (NRDC, 2021). To mitigate global warming, the Paris Agreement was adopted internationally in 2015, and it aims to limit the temperature increase well below 2°C and pursue efforts to limit it to 1.5°C by the end of this century (Masson-Delmotte et al., 2018).

However, based on the “global stocktake” in the latest Conference of the Parties (COP 28) in 2023, the world is still well off-track in securing a 1.5°C word (Zoo, 2023). To achieve the targets of the Paris Agreement, nearly all modeled scenarios show that emissions reduction alone is no longer sufficient (Rau & Greene, 2015), and carbon dioxide removal (CDR) is required by an extensive deployment of negative emission technologies (NETs) (EASAC Policy Report 35, 2018; Johansson et al., 2020). By using NETs, CO$_2$ can be removed from the atmosphere, and they can offset emissions from industries and sectors that are hard to mitigate.

Compared to nature-based NETs, such as afforestation/reforestation, biochar/soil carbon, ocean fertilization, and enhanced weathering, bioenergy with carbon capture and storage (BECCS) is a large-scale engineering solution (Quader & Ahmed, 2017). According to the International Energy Agency, the capability of BECCS could remove 10 GtCO$_2$/year by 2050 (EBTP, 2018). Therefore, BECCS is emerging as an available solution with a large capacity of negative emissions (Bellamy et al., 2019; Bui, Adjiman, et al., 2018; Fajardy et al., 2018).

Integrating CO$_2$ capture with combined heat and power (CHP) plants shows excellent opportunities for the commercial operation of BECCS (Shahbaz et al., 2021). Biomass and waste are commonly used as fuel for CHP plants (Anca-Couce et al., 2021), especially in northern Europe. Therefore, integrating BECCS with biomass-fired and waste-fired CHP (bio-CHP and w-CHP) plants is a promising solution to reduce CO$_2$ emissions. Taking Sweden as a
case study, the CO₂ emission reduction potential is around 20 MtCO₂/year (Beiron et al., 2022).

1.2 Knowledge gaps and challenges

Among different capture technologies, monoethanolamine (MEA) based chemical absorption (MEA-CA) has already been commercialized (Chu et al., 2016; Li et al., 2023). MEA-CA separates CO₂ based on the reversible chemical reaction between CO₂ and the MEA solution. It has been well recognized that the change in the flue gas (FG) entering the CO₂ capture system, such as flowrates and compositions, can clearly affect the performance of CO₂ capture (Li et al., 2019; Tan et al., 2016). Compared to coal-fired power plants, bio/w-CHP plants are characterized by high fluctuations in operation, which further requires a deeper understanding of the dynamic performance of CO₂ capture. First, using versatile biomass and waste as fuel can produce varying FG flowrates and compositions (Malmgren & Riley, 2018). According to the measured data provided by a Swedish utility company, Malarenergi, the CO₂ volume concentration (CO₂vol%) can vary from 8% to 16%, and the flowrates can vary more than 30% during a year (Mälarenergi, 2023). In addition, the bio/w-CHP plants in Nordic countries are usually equipped with back-pressure steam turbines, and the core business is to supply heat to district heating (DH) networks. The operation of CHP plants dynamically varies with the heat demand, which can also lead to dynamic variations in the FG flowrate. Moreover, heat competition exists between heat supply and CO₂ capture. The available heat for CO₂ capture also varies dynamically depending on the heat and electricity generation. Therefore, it is more important to do dynamic simulations for CO₂ capture in bio/w-CHP plants than in coal-fired power plants.

There are different physical model-based approaches for dynamic modeling CO₂ capture, such as using the steady-state model dynamically, using dynamic models without controllers, and using dynamic models with controllers. However, there hasn’t been any guidance on selecting modeling approaches for CO₂ capture in bio/w-CHP plants. Thus, the first research question is:

*RQ1: How should the approach be selected for dynamically modeling CO₂ capture in bio/w-CHP plants?*

The performance of different approaches is clearly influenced by the variation of operating parameters. For bio/w-CHP plants, such parameters include the FG flowrate, the FG CO₂vol%, and the available heat for CO₂ capture. However, there haven’t been any studies about their influences. Thus, the second research question is:
**RQ2: What is the influence of critical operating parameters of bio/w-CHP plants on the performance of different modeling approaches?**

Due to the complexity of physical models, using a detailed physical model is computationally demanding and time-consuming. Data-driven technologies are merging as new methods to replace physical models. However, it has received less attention regarding dynamically modeling CO$_2$ capture. Therefore, the third research question is:

**RQ3: How does the data-driven approach perform when being used for dynamically modeling CO$_2$ capture?**

### 1.3 Scope and objectives

The overall scope of this thesis is to provide guidance on the approach selection for dynamically modeling the MEA-based CO$_2$ capture in bio/w-CHP plants. Corresponding to the above-listed research questions, specific objectives include the following:

- To evaluate the performance of different physical model-based approaches and provide guidance for approach selection (Q1);
- To understand the influence of critical operating parameters from bio/w-CHP plants on the performance of different modeling approaches (Q2);
- To verify the feasibility of the data-driven approach for dynamically modeling CO$_2$ capture (Q3).

### 1.4 Methodology

The methodology adopted in this thesis is illustrated in Figure 1.1. A comprehensive literature survey has been done to learn state-of-the-art dynamic approaches to modeling CO$_2$ capture. Based on the collected approaches, different physical models are implemented in the simulation tool Aspen Plus and HYSYS. To compare the performance of different approaches, an actual w-CHP plant has been chosen as a case study, and various cases have been defined considering different critical operating parameters. Via comparison, suggestions are provided regarding approach selection. To further develop and test the data-driven approach, the time series-based machine learning model, Informer, has been employed, and the data generated from the developed physical model are used for model training and validation.
1.5 Thesis structure

This thesis consists of the following chapters:

Chapter 1  Introduction
This chapter introduces the background, knowledge gaps and challenges, objectives, overall methodology, and thesis outline.

Chapter 2  Literature review
This chapter reviews the dynamic modeling approaches of MEA-based CO₂ capture.

Chapter 3  Approaches for modeling CO₂ capture in bio-CHP plants
This chapter describes the modeling development of different approaches and the defined cases. Both the physical model-based approaches and the data-driven model-based approaches are included.

Chapter 4  Results and discussions
This chapter shows the comparison results of different physical model-based approaches, suggestions about the approach selection, and the performance of the data-driven approach.

Chapter 5  Summary of papers
This chapter summarizes the results of the appended papers and highlights the author’s contributions.

Chapter 6  Conclusions
This chapter highlights the main findings of this thesis.

Chapter 7  Future work
This chapter introduces the planned future work.
2 Literature review

This chapter provides a literature review of previous studies about integrating BECCS in biomass/waste CHP plants (2.1), the physical model-based approaches to CO₂ capture (2.2), which could refer to the Introduction Section in Paper I, and the data-driven approach (2.3), which could refer to the Introduction Section in Paper II. Current research gaps are also summarized at the end.

2.1 Integrating BECCS in biomass/waste CHP plants

For MEA-CA, the FG enters the absorber from the bottom and contacts counter-currently with a lean MEA solution. After absorption, the rich MEA solution is sent into the stripper, in which CO₂ is regenerated from the top when heat duty (100–120 °C steam) is provided to the reboiler, and the resulting lean MEA solution is recirculated back to the absorber (Nittaya et al., 2014; Notz et al., 2012).

Many studies have investigated the technical-economic performance of integrating MEA-CA with bio/w-CHP plants. For example, Pröll and Zerobin assessed the amount of captured CO₂ and its resulting energy penalty of integrating MEA-CA with a 66 MW bio-CHP plant (Pröll & Zerobin, 2019). It was estimated that the captured CO₂ was 1323 kg/(MWh heat). Due to the energy penalty of CO₂ capture, the heat and electricity output of the CHP plant were reduced by 53% and 3%, respectively. Beiron et al. conducted a techno-economic assessment of CO₂ capture from 110 existing Swedish bio-CHP and w-CHP plants (Beiron et al., 2022). It was found that up to 19.3 MtCO₂/year could be captured at 45–125 €/tCO₂ when CO₂ was captured and transported to the nearest harbor by truck. Operating CHP plants with CO₂ capture caused reductions in electricity and DH production levels of around 20% and 40%–60%, respectively.

Most of the previous studies on capturing CO₂ from bio/w-CHP plants are based on steady-state operation. However, as aforementioned, bio/w-CHP plants are characterized by high fluctuations, so dynamic simulations are necessary.
2.2 Physical model-based approaches to CO₂ capture

Modeling the dynamic CO₂ capture by MEA-CA can be done differently. Three approaches commonly used in literature are presented in Table 2.1, as referred to in Table 1 in *Paper I*.

Table 2.1: Three approaches to dynamic modeling of CO₂ capture.

<table>
<thead>
<tr>
<th>Modeling approaches</th>
<th>Using ideal static models (IST) (Martinez Castilla et al., 2019)</th>
<th>Using dynamic models</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mechanism</td>
<td>This approach dynamically uses a steady-state model developed based on chemical and physical equilibrium assumptions. When the steady-state model is used to do dynamic simulations, it is assumed that the equilibrium performance is maintained in each time step, disregarding the time required to reach equilibrium.</td>
<td>To reflect the natural response of CO₂ capture, the dynamic model is developed to simulate the mass transfer, heat transfer, and chemical reactions between gas and MEA solution. The dynamic model shows the change in performance with time. The time needed for establishing equilibrium is considered.</td>
</tr>
<tr>
<td>Advantages</td>
<td>The steady-state model is easy to develop, and the calculating time is short.</td>
<td>The dynamic model can reflect the actual response to input parameter variation.</td>
</tr>
<tr>
<td>Disadvantages</td>
<td>The steady-state model cannot capture the natural dynamic variation of process variables. Its performance largely depends on the time step when used for dynamic simulations. It can be even more time-consuming when a small time step is used.</td>
<td>The calculating time is long, and significant input variations could lead to unstable operation, making it hard for the model to converge.</td>
</tr>
<tr>
<td>Applications</td>
<td>It is commonly used for feasibility studies, system design, and process planning.</td>
<td>It is usually used to study the system response, such as the stability and the response speed, which is essential for controller development.</td>
</tr>
</tbody>
</table>

The steady-state model can be used to do dynamic simulations, called the “ideal static model (IST).” The process is assumed to be steady in each time step, but the input parameters can vary for different time steps. For example, Martinez Castilla et al. developed an IST to study the influence of the variation of available heat for CO₂ capture (Martinez Castilla et al., 2019). Results
showed that, with the time step of 1 hour, the amount of captured CO\textsubscript{2} was 41.1 kton over a two-week period, when the available heat varied hourly in the range of 80-150 MW (average 110 MW), and the FG flowrate was kept constant (140 kg/s).

It is obvious that dynamic models can be used. Without controllers being implemented, dynamic models (Dw/oC) can be used to study the natural response of CO\textsubscript{2} capture in the face of disturbance. For example, Åkesson et al. developed a rate-based dynamic model of MEA-CA using Modelica to study the effect of FG flowrate fluctuation, which was reduced by 30% (Åkesson et al., 2012). The CO\textsubscript{2} removal rate in the absorber was shown to increase rapidly, while more than 1 hour was required for the temperature of the stripper top stage to reach a new steady state. The FG flowrate had little effect on reboiler temperature. Bui et al. developed a dynamic model to gain insight into the interaction between key process parameters in the face of realistic and flexible operation (Bui, Tait, et al., 2018). The results showed that increasing the liquid-to-gas (L/G) ratio (the MEA solution and flue gas) could result in higher CO\textsubscript{2} capture rates. Turning off the heat supply to the reboiler would lead to a gradual decline in reboiler temperature, which will thus increase solvent lean loading and reduce CO\textsubscript{2} capture rate.

Dynamic models with controllers implemented (DwC) can handle the disturbance of input variations. For example, Lin et al. developed a dynamic model with proportional–integral (PI) controllers in Aspen Dynamics (Lin et al., 2011). The results showed that, for the increase of FG flowrate (a step change of 10%) or CO\textsubscript{2} concentration (a step change of 16%), one PI controller was added to adjust the recycle solution flowrate, and a 90% removal target can be maintained. Another PI controller was added to manipulate the reboiler duty to control the reboiler temperature, and a desired lean solution loading of 0.38 molCO\textsubscript{2}/mol MEA can be achieved. Magnanelli et al. developed a dynamic model for MEA-CA in MATLAB to study the performance of a w-CHP plant integrated with MEA-CA (Magnanelli et al., 2021). To achieve an 85% CO\textsubscript{2} capture rate, similar to the study of Lin et al. (Lin et al., 2011), the recycle solution flowrate and reboiler duty needed to be adjusted based on the FG. It was found that capturing 85% of CO\textsubscript{2} can reduce the heat and electricity production by 6% and 30% in a year. However, considering capturing CO\textsubscript{2} seasonally only when there is excess heat, the FG and available heat fluctuations were included. One PI controller was needed to adjust the recycle solution flowrate to achieve the designed L/G ratio of 3 kg/kg. Results showed that reaching a capture rate of 47% was possible, and electricity production was reduced by 5% in a year (Magnanelli et al., 2021).

There are some comparisons of different dynamic modeling approaches. For example, Montañés et al. compared the IST with the DwC with different control structures in Modelica for MEA-CA (Montañés et al., 2017). Results showed that the difference in the amount of captured CO\textsubscript{2} was approximately 2% over 8 hours when the FG flowrate was ramped up from 70 to 100% in 3
minutes. Martinez Castilla et al. estimated the CO\textsubscript{2} captured from an industrial steel mill CHP plant (Martinez Castilla et al., 2019). Four models were compared, including a “steady-state model” with a constant heat load and IST, Dw/oC, and DwC with the actual heat load. Results showed that the amounts over a two-week period were 40.9 kton, 41.1 kton, 41.9 kton, and 42.4 kton for the four models, respectively.

Based on the previous studies, although there are a few comparisons of different modeling approaches, there hasn’t been any guidance on the approach selection. In addition, most studies focus on the parameters’ step change or ramp change, which are not the actual variations from bio/w-CHP plants. Moreover, only the variation of the FG flowrate or the available heat was considered. There is a lack of comprehensive studies evaluating all possible critical operating parameters from bio/w-CHP plants. Furthermore, the time step is another crucial parameter for dynamic simulations, which can clearly affect the model accuracy. However, little attention has been paid to studying its influence.

2.3 Data-driven approaches to CO\textsubscript{2} capture

The detailed physical modeling of CO\textsubscript{2} capture will bring challenges for optimization (Li et al., 2018). To overcome this problem, data-driven models can be developed based on the process data (Wu et al., 2020). However, only a few studies focus on investigating data-driven models. For example, Li et al. (F. Li et al., 2015) developed the bootstrap aggregated neural networks (BA-NN) model for MEA-CA to predict the flowrate of captured CO\textsubscript{2} by employing the following 7 parameters as inputs: the FG flowrate, CO\textsubscript{2}vol%, the FG pressure, the FG temperature, the lean solution flowrate, the MEA concentration and the temperature of the lean solution. The results showed that the mean squared error (MSE) of BA-NN is 0.02 for predicting the flowrate of captured CO\textsubscript{2}.

Most previous studies are based on neural networks. However, the neural network models need to be trained with large, labeled datasets, and the training process is costly and time-consuming.
3 Approaches to modeling dynamic CO₂ capture in bio/w-CHP plants

This chapter provides a methodology for different physical model-based approaches to modeling dynamic MEA-CA (3.1), which could refer to the Appendix A1-A3 Sections in Paper I; defined cases based on an actual w-CHP plant (3.2), which could refer to the Methods Section in Paper I; and the development of data-driven approach (3.3), which could refer to the Methods Section in Paper II.

3.1 Physical model-based approaches to CO₂ capture

To select a proper approach to dynamic modeling of CO₂ capture in bio/w-CHP plants, different approaches should be first developed for further comparison and difference identification. Considering the design, operation, and optimization of the CO₂ capture process, three different models are developed for MEA-CA, including ideal static models, dynamic models without control, and dynamic models with control.

3.1.1 Approach of ideal static model (IST)

When IST is used for dynamic simulations, the input parameter is varied at each time step of 30 minutes, and the new steady state can be attained at the same time as the boundary is changed. For the approach of using IST, a steady state model is first developed in Aspen Plus, with the flowsheet shown in Figure 3.1, as referred to in Appendix A1 (Figure A1) in Paper I. The rate-based model is employed for both the absorber and stripper. The same column parameters as the Tarong post-combustion capture (PCC) pilot plant are used, as shown in Table 3.1 (K. Li et al., 2015), as referred to Appendix A1 (Table A1) in Paper I. Then the steady-state model is validated with the measured data from the Tarong pilot plant (K. Li et al., 2015), which details can be found in Appendix A1.
Table 3.1: Summary of model parameters and column settings (K. Li et al., 2015).

<table>
<thead>
<tr>
<th>Model and column properties</th>
<th>Absorber</th>
<th>Stripper</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of stages</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Packing material</td>
<td>IMTP#38MM</td>
<td>IMTP#38MM</td>
</tr>
<tr>
<td>Total packed height</td>
<td>7.136 m (4x1.784 m)</td>
<td>7.168 m (2x3.584 m)</td>
</tr>
<tr>
<td>Column diameter</td>
<td>350 mm</td>
<td>250 mm</td>
</tr>
<tr>
<td>Flow model</td>
<td>Mixed model</td>
<td>Mixed model</td>
</tr>
<tr>
<td>Interfacial area factor</td>
<td>1.8</td>
<td>1.8</td>
</tr>
<tr>
<td>Initial liquid holdup</td>
<td>0.03 L</td>
<td>0.03 L</td>
</tr>
<tr>
<td>Film resistance</td>
<td>Discrxn for liquid; Film for vapor</td>
<td>Discrxn for liquid; Film for vapor</td>
</tr>
<tr>
<td>Discretization points for liquid film</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>Mass transfer correlation</td>
<td>Bravo et al. (Bravo et al., 1985)</td>
<td>Bravo et al. (Bravo et al., 1985)</td>
</tr>
<tr>
<td>Heat transfer correlation</td>
<td>Chilton-Colburn</td>
<td>Chilton-Colburn</td>
</tr>
<tr>
<td>Interfacial area method</td>
<td>Bravo et al. (Bravo et al., 1985)</td>
<td>Bravo et al. (Bravo et al., 1985)</td>
</tr>
<tr>
<td>Liquid holdup correlation method</td>
<td>Bravo et al. (Rocha et al., 1993)</td>
<td>Bravo et al. (Rocha et al., 1993)</td>
</tr>
</tbody>
</table>

3.1.2 Dynamic approach without control (Dw/oC)

For Dw/oC, a dynamic model is developed, and the time step is 5 seconds. The dynamic model is developed in Aspen HYSYS V12.1, and it is based on the pressure-flow solver, in which the pressure loss is correlated to the flowrate of the streams. The flow diagram is shown in Figure 3.2, and the primary input parameters are shown in Table 3.2, as referred to Appendix A2.
(Figure A2 and Table A4) in Paper I. Dw/oC is validated with the results from Harun et al. (Harun et al., 2012), which details can be found in Appendix A2.

![Diagram of MEA-based CO2 capture without control strategy in Aspen HYSYS.](image)

Figure 3.2: MEA-based CO2 capture without control strategy in Aspen HYSYS.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Absorber</strong></td>
<td></td>
</tr>
<tr>
<td>Packing height and column diameter (m)</td>
<td>6.1/0.43</td>
</tr>
<tr>
<td>Packing type</td>
<td>IMTP#38MM*</td>
</tr>
<tr>
<td>Stage number</td>
<td>10</td>
</tr>
<tr>
<td><strong>Stripper</strong></td>
<td></td>
</tr>
<tr>
<td>Packing height and column diameter (m)</td>
<td>6.1/0.43</td>
</tr>
<tr>
<td>Packing type</td>
<td>IMTP#38MM*</td>
</tr>
<tr>
<td>Stage number</td>
<td>10</td>
</tr>
<tr>
<td><strong>FG, Solution, and Reboiler duty</strong></td>
<td></td>
</tr>
<tr>
<td>CO₂ volume concentration in FG (vol%)</td>
<td>17.5</td>
</tr>
<tr>
<td>Solution flowrate (kg/s)</td>
<td>31.4</td>
</tr>
<tr>
<td>Reboiler duty (kW)</td>
<td>155</td>
</tr>
</tbody>
</table>

* IMTP#38MM means Intalox Metal Tower Packing, with a size specification of 38mm.

3.1.3 Dynamic approach with control (DwC)

For DwC, the same dynamic model in Chapter 3.1.2 is used but with controllers implemented, and the time step is also 5 seconds. Different PI controllers have been integrated to achieve the set targets, as shown in green lines in Figure 3.3, as referred to in Appendix A3 (Figure A5) in Paper I.
In the face of the FG disturbance (FG flowrate and CO₂ vol%), controllers are implemented differently depending on the available heat ($Q_{reb}$). When there is enough heat that can meet the requirement of the reboiler, 7 PI controllers are integrated, as shown in Table 3.3, as referred to in Appendix A3 (Table A7) in Paper I.

Table 3.3: Manipulated and controlled variables and target of controllers (Harun et al., 2012; Nittaya et al., 2014).

<table>
<thead>
<tr>
<th>Controller</th>
<th>Manipulated variable</th>
<th>Controlled variable</th>
<th>Target</th>
</tr>
</thead>
<tbody>
<tr>
<td>FIC1</td>
<td>Lean solution flowrate</td>
<td>Removal rate (when the available heat can meet the need for a reboiler)</td>
<td>90%</td>
</tr>
<tr>
<td>FIC2</td>
<td>Make-up of MEA and H₂O</td>
<td>Mass balance</td>
<td>MEA 30wt%</td>
</tr>
<tr>
<td>TIC1</td>
<td>The cooling supply to the condenser</td>
<td>Condenser T</td>
<td>350K</td>
</tr>
<tr>
<td>TIC2</td>
<td>The heat supply to reboiler</td>
<td>Reboiler T</td>
<td>386K</td>
</tr>
<tr>
<td>TIC3</td>
<td>The cooling supply to the cooler</td>
<td>Lean solution T</td>
<td>314K</td>
</tr>
<tr>
<td>LIC1</td>
<td>Reflux flowrate</td>
<td>Liquid level of condenser</td>
<td>50%</td>
</tr>
<tr>
<td>LIC2</td>
<td>Recycle solution flowrate</td>
<td>Liquid level of reboiler</td>
<td>50%</td>
</tr>
</tbody>
</table>

For example, to maintain the CO₂ removal rate around its nominal set point, the flowrate controller, FIC1, is used by manipulating the lean MEA solution flowrate. The CO₂ removal rate and lean MEA solution flowrate are thus controlled and manipulated variables, respectively. In addition, the CO₂ removal
rate is calculated based on the CO₂ amount in the FG and the CO₂ amount in the vented gas from the top of the absorber, as shown with red lines. To maintain the temperature and the liquid level around their set points, the temperature controllers (TIC) and the liquid level controllers (LIC) are used by adjusting the heat/cooling supply and the outlet solution flowrate, respectively. Using TIC2 could maintain the reboiler temperature and thus keep the lean loading and capture rate by manipulating the reboiler duty (Lin et al., 2011).

However, when the available heat is used as the input parameter (with fixed reboiler duty), there will be no TIC2. Instead of maintaining the removal rate, FIC1 is used to manipulate the lean MEA solution flowrate to keep the lean loading.

3.2 Case study

Comparison and difference identification are crucial to recommendations on the approach selection. To compare different approaches, an actual w-CHP plant is chosen as an example to enable the same input. Models are scaled up based on the capacity of the w-CHP plant. In addition, considering different operating parameters of bio/w-CHP plants, various cases are defined to find out their influences on different approaches. Key performance indicators are also defined at the end for comparison.

3.2.1 Data from the CHP plant and model scale-up

The FG data collected from an actual w-CHP plant from 2017/1/1 to 2017/12/31 are used as input parameters for simulations and are illustrated in Figure 3.4, as referred to in Section 2.2.1 (Figure 1) in Paper I. The thermal capacity of the plant is 167 MWth, and the fuel mainly consists of household waste, industrial waste, and recycled wood. The time resolution is 30 minutes. In general, the variation of FG flowrate and CO₂vol% are in the range of 180.57-375.21 kNm³/h and 7.70-15.72vol%, respectively.

The models presented in Chapter 3.1 are developed and validated under different FG flowrates. Therefore, they need to be scaled up to meet the requirements of the studied w-CHP plant. Based on the method proposed by Otitoju et al. (Otitoju et al., 2020), the dimensions of the absorber and stripper are estimated, including the diameter and height of the packed columns, the details of which can be found in Appendix A3. For other vessels, including the reboiler and the buffer tank between the absorber and stripper, their volumes are determined based on a residence time of 10 minutes for the incoming liquid and a liquid level of 50%. The results after the model scale-up are shown in Table 3.4, as referred to in Appendix A1-A3 (Table A3, A6, and A8) in
\textit{Paper I.} The other needed input parameters are also included in Table 3.4, including the lean MEA solution and FG.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fg_data.png}
\caption{FG data from a w-CHP plant.}
\end{figure}

\begin{table}[h]
\centering
\begin{tabular}{lll}
\hline
\textbf{Components} & \textbf{Value} & \textbf{Input Streams} & \textbf{Value} \\
\hline
\textbf{Absorber} & & Lean MEA Solution & \\
Packing type & IMTP\#38MM & MEA concentration (wt\%) & 30 \\
Packing height and column diameter (m) & 17.4/8.7 & Lean loading & 0.287 \\
Pressure (kPa) & 101.3 & Solution temperature (K) & 314 \\
\textbf{Stripper} & & Solution flowrate (kg/h) & \\
Packing type & IMTP\#38MM & FG & \\
Packing height and column diameter (m) & 10.9/4.3 & FG temperature (K) & 319.7 \\
Reboiler volume (m3) & 352 & O₂ (vol\%) & 2.7 \\
The volume of the tank between the absorber and stripper (m3) & 330 & H₂O (vol\%) & 4.8 \\
Condenser/reboiler temperature (K) & 350/386 & & \\
Condenser pressure (kPa) & 159 & & \\
\hline
\end{tabular}
\caption{The scaled-up capture system and other input parameters for the case study.}
\end{table}
3.2.2 Critical operating parameters

Various cases are defined based on the critical operating parameters from the w-CHP plant. For situations where no actual data can be obtained, fabricated data are used as supplementary data based on the assumption of sinusoidal changes, which will be described later. The critical operating parameters considered in this work include the FG flowrate, the FG CO₂vol%, and the available heat for CO₂ capture. To study their influences, 5 cases are defined: Case 1, in which the FG flowrate varies (Figure 3.5); Case 2, in which the CO₂vol% of FG varies (Figure 3.6); Case 3, in which both the FG flowrate and CO₂vol% vary (Figure 3.7); Case 4, in which the available heat for CO₂ capture varies (Figure 3.8); and Case 5, in which both FG (the flowrate and CO₂vol%, same as Case 3) and available heat vary (Figure 3.9). Figures 3.5 to 3.9 can be referred to in Section 2.2.2 (Figure 2-6) in Paper I.

For Case 1 and Case 3, actual FG flowrate data selected from the example w-CHP plant operation are used. For Case 2, it is difficult to find a period in which only the CO₂vol% varies but not the flowrate; therefore, sinusoidal changes in a range of 9.7-15.7vol% are assumed for CO₂vol%. For Cases 1 to 3, the available heat is assumed to meet the reboiler heat demand for capturing 90% of CO₂. For Case 4, since the example plant doesn’t include CO₂ capture, the available heat for CO₂ capture cannot be obtained directly from measurements. Therefore, it is also assumed that the available heat changes sinusoidally in the range of 49-96 MW. For Case 5, the data same as for Case 3 are used for the FG flowrate and CO₂vol%, and the available heat is assumed to change sinusoidally in a range of 90-104 MW. Because Harun found that the decrease and increase of the FG flowrate could affect CO₂ capture operation differently (Harun, 2012), each case is further divided into two sub-cases, corresponding to the increasing and decreasing variations. Considering the workload for calculation, simulations are performed for 4 hours. Case 1 to Case 5 are also compared in Table 3.5, as referred to in Section 2.2.2 (Table 2) in Paper I.

![Figure 3.5: Variation of FG flowrate in Case 1.](image-url)
Figure 3.6: Variation of CO$_2$vol% in Case 2.

Figure 3.7: Variation of FG flowrate and CO$_2$vol% in Case 3.

Figure 3.8: Variation of the available heat in Case 4.

Figure 3.9: Variation of the available heat in Case 5.
Table 3.5: An overview of the defined cases.

<table>
<thead>
<tr>
<th>Case</th>
<th>Parameters</th>
<th>Sub-cases</th>
<th>Detailed data</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>FG flowrate *</td>
<td>Case 1-1 (decrease)</td>
<td>FG flowrate:</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Case 1-2 (increase)</td>
<td>Case 1-1: from 340.77kNm³/h to 264.73kNm³/h (91% to 70%) *</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 1-2: from 201.68kNm³/h to 337.02kNm³/h (53% to 90%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>CO₂vol%: 14.4% (average)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Available heat: heat can meet the reboiler heat demand for a CO₂ capture rate of 90%</td>
</tr>
<tr>
<td>Case 2</td>
<td>FG CO₂vol% b</td>
<td>Case 2-1 (decrease)</td>
<td>FG flowrate:</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Case 2-2 (increase)</td>
<td>Case 2-1: from 15.7% to 9.7% (100% to 61%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 2-2: from 9.7% to 15.7% (61% to 100%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Available heat: heat can meet the reboiler heat demand for a CO₂ capture rate of 90%</td>
</tr>
<tr>
<td>Case 3</td>
<td>FG flowrate a</td>
<td>Case 3-1 (flowrate decrease and CO₂vol% increase)</td>
<td>FG flowrate:</td>
</tr>
<tr>
<td></td>
<td>+ CO₂vol% b</td>
<td>Case 3-2 (flowrate increase and CO₂vol% decrease)</td>
<td>Case 3-1: from 375.21kNm³/h to 326.14kNm³/h (100% to 86%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 3-2: from 327.94kNm³/h to 351.00kNm³/h (87% to 94%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>CO₂vol%: (Sinusoidal variation)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 3-1: from 12.0% to 13.6% (76% to 87%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 3-2: from 13.4% to 12.4% (85% to 79%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Available heat: heat can meet the reboiler heat demand for a CO₂ capture rate of 90%</td>
</tr>
<tr>
<td>Case 4</td>
<td>Available heat</td>
<td>Case 4-1 (decrease)</td>
<td>FG flowrate:</td>
</tr>
<tr>
<td></td>
<td>for CO₂ capture b</td>
<td>Case 4-2 (increase)</td>
<td>Case 4-1: from 96MW to 49MW</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 4-2: from 49MW to 96MW</td>
</tr>
<tr>
<td>Case 5</td>
<td>FG flowrate a</td>
<td></td>
<td>FG: same as Case 3-1 and 3-2</td>
</tr>
<tr>
<td></td>
<td>+ CO₂vol% b</td>
<td></td>
<td>Case 5-1: from 375.21kNm³/h to 326.14kNm³/h (100% to 86%)</td>
</tr>
<tr>
<td></td>
<td>+ available heat b</td>
<td></td>
<td>Case 5-2: from 327.94kNm³/h to 351.00kNm³/h (87% to 94%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>CO₂vol%: (Sinusoidal variation)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 5-1: from 12.0% to 13.6% (76% to 87%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Case 5-2: from 13.4% to 12.4% (85% to 79%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Available heat: (Sinusoidal variation)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>from 90MW to 104MW first and then from 104MW to 90MW</td>
</tr>
</tbody>
</table>

* It means the actual data from the CHP plant. b It means the fabricated data. * The value in brackets means their percentage compared to the maximum value of actual data.

For all approaches, the lean MEA solution flowrate and the reboiler duty (available heat) are two critical parameters. When IST and DwC are used, if the available heat can always meet the reboiler’s requirement (Case 1 to Case
3), the lean MEA solution flowrate and reboiler duty are adjusted accordingly to achieve the set target of CO₂ capture rate (90%), according to FG flowrate and CO₂vol%. When the available heat is specified (Case 4 and Case 5), for IST, the captured CO₂ is calculated based on the available heat and specific heat needed by capturing 1 kg CO₂, while for DwC, the controller will regulate the lean MEA solution flowrate to achieve the set target of lean loading (0.287 mol/mol), according to FG flowrate, CO₂vol% and the available heat. When Dw/oC is used, if the available heat can always meet the reboiler’s requirement (Case 1 to Case 3), lean MEA solution flowrate and reboiler duty remain constant, which are determined based on the average FG CO₂ content and a fixed capture rate (90%). When the available heat is specified (Case 4 and Case 5), the lean MEA solution flowrate remains constant, which is determined based on the average available heat and fixed lean loading (0.287 mol/mol).

### 3.2.3 Key performance indicators

The accumulated amount of captured CO₂ and the average specific reboiler duty are key performance indicators (KPIs) to compare different approaches. The amount of captured CO₂ is calculated by integrating dynamic results over 4 hours, as shown in Equation 3-1. The average specific reboiler duty is calculated by taking the average over 4 hours of specific reboiler duty, as shown in Equation 3-2. The equations can be referred to in Section 2.2.2 (Eqn.1-2) in Paper I.

\[
\text{Captured CO}_2 = \int_0^T \text{CO}_2\text{pro}(t) \, dt \quad \text{(3-1)}
\]

\[
\text{Average specific reboiler duty} = \frac{\int_0^T Q_{reb}(t) \, dt}{\int_0^T \text{CO}_2\text{pro}(t) \, dt} \quad \text{(3-2)}
\]

where \(\text{CO}_2\text{pro}(t)\) is the amount of CO₂ in the product stream at the time \(t\), ton/h; \(\text{Captured CO}_2\) is the accumulated amount of captured CO₂ over 4 hours, ton/4h; \(Q_{reb}(t)\) is the reboiler duty at the time \(t\), GJ/h; and \(\text{Average specific reboiler duty}\) is the average heat penalty per unit captured CO₂ over 4 hours, MJ/kg.

### 3.3 The data-driven approach

To study how the data-driven approach performs in dynamically modeling CO₂ capture, a time series machine learning method, Informer, is selected. The dataset is generated by using the developed physical model in Chapter 3.1.
3.3.1 Dataset preparation

The developed MEA-CA model, Dw/oC, in the Aspen HYSYS simulator, is used to generate the dataset. The CO₂ capture rate \( y_1 \) and the energy penalty per unit captured CO₂ \( y_2 \) are two key indicators to show the operating performance of CO₂ absorption and desorption. Therefore, they are chosen as output parameters. The following essential parameters are selected as inputs, including FG flowrate \( u_1 \), FG CO₂ concentration \( \text{CO}_2\text{vol\%} \) \( u_2 \), the lean solvent flowrate \( u_3 \), and the reboiler heat input (or the available heat for CO₂ capture) \( u_4 \), as shown in Figure 3.10.

The random function generates the excitation signal of 4 input parameters with step change every 15 minutes, as shown in Figure 3.11, as referred to in Section 2.2 (Figure 2) in Paper II. Based on the FG flowrate and FG CO₂ vol%, the reboiler heat input and solution flowrate ranges are thus determined by the assumption of a 50-90% capture rate. Based on the excitation signal in Figure 3.11, the dynamics of CO₂ capture are simulated using the Dw/oC of the MEA-CA model. As outputs, CO₂ capture rate and energy penalty are calculated based on Equations 3-3 and 3-4, as referred to in Section 2.2 (Eqn.1-2) in Paper II. All input and output data are sampled every 5 s to form a dataset of 12,060 groups of sampled data within 1005 mins.

\[
\text{CO}_2 \text{ capture rate} = \frac{\text{CO}_2\text{pro}}{\text{CO}_2\text{FG}} \times 100\% \quad (3-3)
\]

\[
\text{Energy penalty} = \frac{Q_{reb}}{\text{CO}_2\text{pro}} \text{ (kJ/kg CO}_2) \quad (3-4)
\]

where \( \text{CO}_2\text{pro} \) and \( \text{CO}_2\text{FG} \) are the amounts of CO₂ in the product stream and the FG stream, respectively, ton/h, and \( Q_{reb} \) is the reboiler duty, MJ/h.
3.3.2 The architecture of Informer

In many cases, transformers replace convolutional and recurrent neural networks and become the most popular deep learning models using advanced encoder-decoder structures (Vaswani et al., 2017). The Informer model was proposed to be beyond the efficient Transformer model for long-term time series forecasting (LTSF) tasks (Zhou et al., 2021), which can efficiently capture precise long-term temporal dependency and short-term temporal trends. It shows three distinctive characteristics: (i) a ProbSparse self-attention mechanism lowers time complexity, reduces memory usage, and performs well on sequences' dependency alignment. (ii) the self-attention distilling highlights dominating attention by halving cascading layer input and efficiently handles extremely long input sequences. (iii) the generative style decoder drastically
improves the inference speed of long-sequence predictions by the prediction at one forward operation. Therefore, the Informer model is selected.

This study implements the Informer model in Python, and the PyTorch deep learning framework is used to train the model. The generated data are split into training data (60%), validation data (20%), and testing data (20%). The data are scaled to zero mean and standard deviation variance before being used for model training. Because the temporal complexity of MEA-CA by Aspen HYSYS is not so much, the self-attention layers in the encoder and decoder are set separately as 1 to avoid overfitting. The dimension of feature extraction of layers is 256. The dimension of the last fully connected layer for output generation is 2048. The dropout rate is 0.1. The learning rate is 0.0001. Also, the patience for early stopping is 10 epochs, which means the training process will end if the validation loss does not decrease in the continuous 10 epochs.
4 Results and discussions

This chapter shows the results of the comparison of different physical model-based approaches (4.1), which could refer to Section 3 in Paper I; the approach selection (4.2), which could refer to Section 4 in Paper I; and the performance of Informer (4.3), which could refer to the Results Section in Paper II.

4.1 Comparison on CO₂ capture performance

The dynamic change of KPIs is first shown. Then results of the overall performance are discussed. The influence of the time step is also studied.

4.1.1 Case 1: Variation of the FG flowrate

Figure 4.1 shows the dynamic variations of KPIs in Case 1, as referred to in Section 3.1 (Figure 7) in Paper I. All approaches show that the amount of captured CO₂ varies in the same way as the FG flowrate, as shown by the blue line. It is clear that IST and DwC result in similar variations of the captured CO₂ since both target the same capture rate of 90%. Compared to IST, DwC can lead to a higher (lower) amount of captured CO₂ than IST when the FG flowrate decreases (increases). This is because the variation of the results of DwC is smaller than that of IST. In other words, when the FG flowrate decreases (increases), the reboiler duty needs to be reduced (increased) as less (more) CO₂ will be captured; however, there is a delay in the decrease (increase) of reboiler duty and as a result, the reduction (increase) in CO₂ capture is less than in the case where the equilibrium is reached. For Dw/oC, as there is no control of the lean MEA flowrate and the reboiler duty, they remain constant, which are determined based on the average FG flowrate and 90% capture rate. Taking Case 1-1 as the example, the FG flowrate is higher than the average at the beginning but lower at the end. When the flowrate is higher than the average, even though more CO₂ exists, due to the constant lean MEA flowrate and reboiler duty, not all CO₂ can be captured, which means the actual capture rate is less than 90%. When the flowrate is lower than the average, even though less CO₂ exists, due to the constant lean MEA flowrate and reboiler duty, more CO₂ can be captured, which means an actual capture rate...
higher than 90%. Hence, Dw/oC shows less variation. For Case 1-2, it will be the opposite.

Figure 4.1: Model comparison under the variations of FG flowrate.

For the dynamic specific reboiler duty shown by the orange line, on the contrary, Dw/oC results in the most variation and gives a different trend from DwC and IST. When Dw/oC is used, since the reboiler duty is fixed, when less (more) CO₂ is captured, the specific reboiler duty would increase (decrease) in Case 1-1 (Case 1-2). Compared to the increase in Case 1-1, the decrease in Case 1-2 is more prominent, which is related to how significant the FG flowrate variations are, as shown in Table 3.5. However, when DwC is used, the reboiler duty is adjusted to follow the FG flowrate variations with a time delay. Such a delay can result in more (less) CO₂ being regenerated, which leads to a decrease (increase) in the specific reboiler duty. When IST is
used, the change in the specific reboiler duty is very small as the CO$_2$vol% mainly determines it.

### 4.1.2 Case 2: Variation of the CO$_2$vol%

Figure 4.2 shows the dynamic variations of KPIs in Case 2, as referred to in Section 3.2 (Figure 8) in *Paper I*. Similar to the influence of the FG flowrate in Case 1, all approaches show that the amount of captured CO$_2$ varies in the same way as CO$_2$vol%. Dw/oC shows less variation than IST and DwC; the results of DwC vary more slowly than those of IST. For the dynamic specific reboiler duty, Dw/oC still leads to the most variation due to the fixed reboiler duty. Because a higher CO$_2$vol% can favor the chemical reaction, results of DwC show the same trends as Dw/oC on the specific reboiler duty that increases (decreases) with the decrease (increase) of CO$_2$vol%. For IST, although CO$_2$vol% can affect the specific reboiler duty in the range of 9 - 16%, the effect is small for steady-state simulations (Li et al., 2011).
Figure 4.2: Model comparison under the variations of FG CO$_2$vol%.

4.1.3 Case 3: Variation of both FG flowrate and CO$_2$vol%

Figure 4.3 shows the dynamic variations of KPIs in Case 3, as referred to in Section 3.3 (Figure 9) in Paper I. When combining the FG flowrate and CO$_2$vol%, the influences are determined by the CO$_2$ content, which is the product of FG flowrate and CO$_2$vol%. Therefore, the changes in KPIs don’t follow the changes in FG flowrate and CO$_2$% clearly. When the FG flowrate decreases (increases) while CO$_2$vol% increases (decreases), their impacts on CO$_2$ content become less. In general, DwC and IST give similar results for the captured CO$_2$, which are lower than that of Dw/oC. For the specific reboiler duty, the differences between all approaches are small, less than 6%.
4.1.4 Case 4: Variation of the available heat

Figure 4.4 shows the dynamic variations of KPIs in Case 4, as referred to in Section 3.4 (Figure 10) in Paper I. All approaches show that the amount of captured CO$_2$ varies in the same way as the available heat. Different from the influences of the FG in Case 1 and Case 2, Dw/oC results in more variations than DwC and IST. The variation of the available heat can result in changes in regenerated CO$_2$ and lean loading, which will lead to the variation in the amount of removed CO$_2$ in the absorber and the amount of the captured CO$_2$ in the stripper. When Dw/oC is used, with the decrease (increase) of the available heat, the lean loading increases (decreases). So, the absorption capacity of the lean solution decreases (increases), which, in turn, accelerates the decrease (increase) of the amount of captured CO$_2$. However, when DwC and IST are used, the control of the lean loading is included, which can regulate the lean MEA flowrate according to the available heat to minimize the fluctuation in the absorption.

For the dynamic specific reboiler duty, Dw/oC also shows the most variation, giving an opposite trend to DwC and IST. When Dw/oC is used, the faster decrease (increase) in the amount of captured CO$_2$ will increase (decrease) the specific reboiler duty. However, for DwC and IST, since the regulated lean loading helps minimize the fluctuation in the absorption of lean solution, the difference in the specific reboiler duty is influenced more by the MEA lean solution flowrate ratio to the FG flowrate (L/G ratio). As shown in Table 3.3, the lean MEA flowrate is regulated to achieve the lean loading (0.287 mol/mol). Therefore, when the available heat decreases (increases) to achieve the target lean loading, the lean MEA flowrate is reduced (increased),
which leads to a decrease (increases) in the L/G ratio and a decrease (increase) in the specific reboiler duty.

Figure 4.4: Model comparison under the variations of available heat.

4.1.5 Case 5: Variation of the FG and available heat

Figure 4.5 shows the dynamic variations of KPIs in Case 5, as referred to in Section 3.5 (Figure 11) in Paper I. When combining the FG flowrate, CO2vol%, and the available heat, the influences are determined by both the CO2 content and the available heat. Since the FG flowrate and CO2vol% vary oppositely, which implies the CO2 content doesn’t vary clearly, the changes of KPIs are affected more by the change of the available heat. Therefore, DwC
and IST show a similar increase and decrease of captured CO$_2$ and specific reboiler duty, while Dw/oC shows most variations, similar to Case 4.

![Figure 4.5: Model comparison under the variations of FG and available heat.](image)

### 4.1.6 Comparison on the overall performance

The accumulated amount of captured CO$_2$ and the average specific reboiler duty are shown in Table 4.1, as referred to in Section 3.6 (Table 3) in *Paper I*. The relevant difference is calculated based on the result of IST for Dw/oC and DwC. The differences depend on the magnitude of variations, which means that the more significant the variation of parameters is, the larger the difference is. Dw/oC results in a more substantial difference from the other two approaches. Generally speaking, for the variation of FG flowrate/CO$_2$vol%, DwC will result in lower amounts of captured CO$_2$ than IST with an increased
FG flowrate/CO$_2$vol% and vice versa. In contrast, it is the opposite for the variation of available heat. For Dw/oC, the difference from IST is, to a major degree, dependent on the fact that larger variations lead to larger deviations. For the average specific reboiler duty, compared to IST, DwC shows lower ones except in cases where the available heat decreases, but there is no clear rule for Dw/oC.

Table 4.1: Comparison of different approaches for overall performance.

<table>
<thead>
<tr>
<th>Cases</th>
<th>Amount of captured CO$_2$ (ton/4h)</th>
<th>Average specific reboiler duty (MJ/kg CO$_2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>IST</td>
<td>DwC</td>
</tr>
<tr>
<td>Case 1-1</td>
<td>310.5</td>
<td>+3.9%</td>
</tr>
<tr>
<td>Case 1-2</td>
<td>266.6</td>
<td>-7.2%</td>
</tr>
<tr>
<td>Case 2-1</td>
<td>266.8</td>
<td>+7.3%</td>
</tr>
<tr>
<td>Case 2-2</td>
<td>266.8</td>
<td>-6.5%</td>
</tr>
<tr>
<td>Case 3-1</td>
<td>316.8</td>
<td>+0.2%</td>
</tr>
<tr>
<td>Case 3-2</td>
<td>305.4</td>
<td>-0.1%</td>
</tr>
<tr>
<td>Case 4-1</td>
<td>236.9</td>
<td>-2.4%</td>
</tr>
<tr>
<td>Case 4-2</td>
<td>236.9</td>
<td>+3.6%</td>
</tr>
<tr>
<td>Case 5-1</td>
<td>304.2</td>
<td>+2.1%</td>
</tr>
<tr>
<td>Case 5-2</td>
<td>302.7</td>
<td>+1.4%</td>
</tr>
</tbody>
</table>

4.1.7 Influence of time step (TS)

TS can clearly impact the results of different approaches. To understand such impacts, simulations are performed for Case 5 with TS varied from 1min to 30min for IST and from 0.5s to 5s for Dw/oC and DwC. Figure 4.6 shows the dynamic changes of KPIs, as referred to in Section 3.7 (Figure 12-13) in *Paper I*. It is clear that DwC is more sensitive to TS than Dw/oC and IST. When TS varies from 0.5s to 5s, the maximum variation can be up to 3.4% for the captured CO$_2$ per minute and 3.5% for the specific reboiler duty. This is because controllers take time to regulate the processes, and different designs of controllers have different characters. For Dw/oC, the influence is less evident due to the lack of a controller. For IST, apparent variations in the amount of captured CO$_2$ can also be observed when TS is increased from 5 minutes to 30 minutes because IST gives the accumulated result. Nevertheless, the difference becomes very small when the TS is smaller than 5 minutes. For the specific reboiler duty, TS has negligible impact on the IST because the specific reboiler duty is mainly determined by CO$_2$%.

The accumulated amount of captured CO$_2$, shown in Figure 4.7, as referred to in Section 3.7 (Figure 14) in *Paper I*, can clearly show the influence of TS.
When TS is increased from 0.5s to 5s, the captured CO₂ is decreased by 2.3% (2.6%) for DwC and 0.5% (0.8%) for Dw/oC. For IST, when TS is increased from 1min to 30min, the captured CO₂ is decreased by 0.1% (0.1%). In addition to the captured CO₂, TS also clearly affects the simulation time consumed. For example, by using a laptop with an Intel Core i7-10750H Processor (CPU @ 2.60GHz) and 32GB of installed RAM, when the TS is decreased from 5s to 0.5s, the consumed time of DwC for simulating 4 hour-operation in Aspen HYSYS is increased from around 2 hours to over 8 hours; while, when the TS is decreased from 30min to 1min, the consumed time of IST is increased from 1 minute to around 30 minutes. It is essential to balance the accuracy and the simulation time.

![Figure 4.6: Influence of time step on dynamic changes of KPIs in Case 5-1.](image)

Figure 4.6: Influence of time step on dynamic changes of KPIs in Case 5-1.
4.2 Selection of modeling approaches

The selection of modeling approaches is mainly dependent on the applications. For example, for the equipment design of CO\(_2\) capture, it is essential to check the boundary of safety operation by doing step response analysis and stability analysis. Then, Dw/oC can be chosen. For the control system design of CO\(_2\) capture, the controller needs to be tested to see if it can handle the disturbance and maintain the system’s stability. Hence, DwC should be selected.

For determining the amount of captured CO\(_2\), depending on the input parameters and requirements, DwC and IST may be used. For short-term operations, DwC is suggested, which can consider the impact of non-equilibrium. And the deviation can be up to 7.3%. Nevertheless, for long-period operations, both increasing and decreasing variations are included; the difference between DwC and IST may be small because the positive deviations and negative deviations can cancel each other. One example is illustrated in Figure 4.8, as referred to in Section 4 (Figure 15) in Paper I. The FG flowrate and CO\(_2\)vol% data are taken from the same w-CHP plant. Figure 4.9 compares the captured CO\(_2\), as referred to in Section 4 (Figure 16) in Paper I. Although apparent differences can be observed between the captured CO\(_2\) for each hour, the difference in the daily amount of captured CO\(_2\) is only 5.2 tons, corresponding to a deviation of 0.32%. Considering the more time needed for the model development and running simulations, IST is preferable, and a time step of 30 minutes is recommended.

For the dynamic operation of the CHP plant, the amount of steam that needs to be extracted from the steam turbine cycle should be determined in real-time. For such a case, using either DwC or IST is also possible. Even though the difference is usually less than 5%, DwC is recommended. This is because IST
estimates the heat based on the assumption that the specific reboiler duty depends only on CO\textsubscript{2}vol\%. In contrast, DwC estimates the heat according to the target temperature set to the reboiler. For the solvent regeneration, the temperature has to reach a certain level.

For optimizing the operation of plants integrated with CO\textsubscript{2} capture, it is also possible to use either DwC or IST. The operation optimization is usually carried out based on the prices of electricity and CO\textsubscript{2}. When hourly prices from actual electricity and carbon markets are used, DwC shall be selected. This is because the operation of CO\textsubscript{2} capture can never reach an equilibrium with hourly changes. However, if other price mechanisms are adopted, such as time of use (TOU) or power purchase agreement (PPA), since the prices don’t vary hourly, IST could show advantages over DwC and a big-time step, such as an hour, can be chosen.

![Figure 4.8: FG flowrate and CO\textsubscript{2}vol\% of DwC and IST of one day.](image)

Figure 4.8: FG flowrate and CO\textsubscript{2}vol\% of DwC and IST of one day.
4.3 Performance of Informer model

The mean absolute percentage error (MAPE), defined by Equation 4-1, is used to evaluate the performances.

\[
MAPE = \frac{1}{n} \sum_{i=1}^{n} \left| \frac{y_i - \hat{y}_i}{\bar{y}_i} \right| \times 100\%
\]  

(4-1)

where \( n \) is the sample size, \( y_i \) is the \( i^{th} \) predicted value, and \( \hat{y}_i \) is the \( i^{th} \) actual value.

As shown in Figure 4.10, by using the AI model of Informer, the MAPE is 6.2% for the prediction of CO2 capture rate. In addition, the Informer can portray the character of CO2 capture rate well over the range of 60-100%, but higher deviations over the range below 50% and above 125%. As shown in Figure 4.11, the MAPE of Informer is 2.7% for the prediction of energy penalty. The figures can be referred to in Section 3 (Figure 3-4) in Paper II.
Figure 4.10: The result of CO₂ capture rate.

Figure 4.11: The result of the energy penalty.
5 Summary of papers

This chapter summarizes the main results of each paper appended to this licentiate thesis, highlighting the author’s and co-authors’ contributions. I was the first author of all appended papers, performing most of the writing. My principal supervisor, Prof. Hailong Li, and co-supervisor, Prof. Eva Thorin and Dr. Jan Skvaril, reviewed and commented on the journal paper, Paper I, from the beginning of proposing research questions to the end of the publication.

5.1 Paper I


5.1.1 My contribution

I was the lead author of the paper. I conducted a literature review and identified the approaches for dynamically modeling CO\textsubscript{2} capture. I also developed the models, defined the cases, and performed the simulation and result analysis.

5.1.2 Results and discussions

Paper I compares three physical model-based approaches, namely using the ideal static model (IST) and using dynamic models without control (Dw/oC) and with control (DwC) for dynamically modeling MEA-based chemical absorption CO\textsubscript{2} capture. The performance of approaches is compared under the consideration of the variations of critical parameters, including the flowrate and CO\textsubscript{2}vol\% of FG and the available heat for CO\textsubscript{2} capture. Simulation results show apparent differences. However, when there are both increases and decreases in the variations of parameters, the differences become smaller. It is also found that the time step size clearly impacts the CO\textsubscript{2} capture amount, especially for DwC.
Based on the results, suggestions are also provided regarding the approach selection for different purposes of simulations. It depends on the requirements of the applications. Dw/oC is recommended for checking the boundary of safety operation by the response analysis. DwC is recommended for designing the control system, observing the flexible dynamic operation, estimating the short-term CO$_2$ capture potential, and optimizing the hourly dynamic operation. IST is recommended for estimating the long-term CO$_2$ capture potential, and optimizing the long-term dynamic operation when the input parameters vary not as often as hourly.

5.2 Paper II


5.2.1 My contribution

I was the lead author of the paper. I first prepared the dataset for data-driven modeling. I also determined the input data and conducted model training and validation. Jinyu Chen from the University of Tokyo contributed to training the data-driven modeling by using Informer.

5.2.2 Results and discussions

Paper II aims to develop a data-driven model, Informer, to predict the dynamic responses of MEA-based CO$_2$ capture performance from waste-fired CHP plants. The results verify the feasibility of the data-driven approach for dynamically modeling CO$_2$ capture. It was found that Informer could predict the CO$_2$ capture rate and the energy penalty with the mean absolute percentage error of 6.2% and 2.7%, respectively, when the following variables were used as inputs: the inlet FG flowrate, the CO$_2$ concentration in inlet FG, the lean solvent flowrate, and the heat input to CO$_2$ capture.
Dynamic modeling of CO\textsubscript{2} capture is essential for assessing, controlling, and optimizing CO\textsubscript{2} capture in biomass/waste CHP plants. Three different physical model-based approaches are compared for dynamically simulating CO\textsubscript{2} capture using MEA-based chemical absorption, namely the ideal static model (IST), the dynamic approach without control (Dw/oC), and the dynamic approach with control (DwC). The data-driven model, Informer, has also been established to predict the dynamic responses of MEA-based CO\textsubscript{2} capture. The main conclusions of this thesis are the following:

- Clear differences can be identified amongst approaches. For example, when the CO\textsubscript{2}vol\% drops from 15.7\% to 9.7\% (about 38\%) within 4 hours, the difference in the amount of captured CO\textsubscript{2} can be up to 22\%, which is between DwC and Dw/oC. The difference is also affected by operating parameters. Compared with IST, for the variation of FG flowrate/CO\textsubscript{2}vol\%, DwC results in lower amounts of captured CO\textsubscript{2} with an increased FG flowrate/CO\textsubscript{2}vol\% and vice versa, while for the variation of available heat, it is the opposite. Regarding the average specific reboiler duty, DwC shows lower values than IST except where the available heat decreases.

- When there are both increases and decreases in the variations of parameters, the difference amongst approaches becomes smaller. For the decrease of FG flowrate by 13.5\% and increase of CO\textsubscript{2}vol\% by 13.5\% within 4 hours, the relative difference in the captured CO\textsubscript{2} between DwC and IST is 0.1\%.

- For the approach selection, Dw/oC is recommended for checking the boundary of safety operation; DwC is recommended for designing the control system, observing the flexible dynamic operation, estimating the short-term CO\textsubscript{2} capture potential, and optimizing the hourly dynamic operation; IST is suggested for estimating the long-term CO\textsubscript{2} capture potential, and optimizing the long-term dynamic operation when the input parameters vary not as often as hourly.
The Informer model can predict dynamic CO₂ capture accurately. By employing the following parameters as inputs, the FG flowrate, CO₂vol%, the lean solvent flowrate, and the heat input to CO₂ capture, it was found that the MAPE of Informer is 6.2% and 2.7% for the prediction of CO₂ capture rate and energy penalty, respectively.
7 Future work

Some limitations to the thesis work exist. The 4-hour timescale is determined randomly considering the workload of calculations. It may be interesting to compare the approaches over an extended period. Moreover, only the influence of time steps is investigated. Finding the optimal time step is suggested for future work. Further, to improve the data-driven modeling for future applications, it is suggested to use real operation data from a CO2 capture plant instead of using simulated data.

Besides addressing the limitations, advanced controllers are needed, with the increasing demand for flexible operations of CO2 capture in biomass/waste CHP plants. Model predictive control (MPC) is a commonly applied advanced control technique with the advantages of online optimization, quick regulation, and multi-parameter control. In the future, an MPC will be developed.
References


strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty. Issue.


Forecasting The Thirty-Fifth AAAI Conference on Artificial Intelligence (AAAI-21),
A1: Validation of the steady state model in Aspen plus

The validation for the steady-state model is based on the Tarong pilot plant in Queensland, Australia, in which a PCC plant was constructed with a designed CO₂ capture rate of 85% (around 100 kg/h) using MEA solution based on the coal-fired power station (900 kg/h typical FG flowrate). Different trials have been conducted, and their detailed conditions can be found in Li et al. (K. Li et al., 2015), which are helpful to model development and validation.

As shown in Table A1, including both the input and the output parameters. The maximum deviations are 4.1%, 2.6%, 2.9%, 0.3%, and 0.4% for the CO₂ absorption rate, reboiler temperature, reboiler duty, CO₂ product purity, and CO₂ capture rate, respectively.

Table A1. Comparison between pilot plant trials and steady-state model results (K. Li et al., 2015).

<table>
<thead>
<tr>
<th>Test conditions</th>
<th>Test 1</th>
<th>Test 2</th>
<th>Test 3</th>
<th>Test 4</th>
<th>Test 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Input parameters</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lean temp. °C</td>
<td>31.7</td>
<td>33.9</td>
<td>31.4</td>
<td>31.3</td>
<td>35.5</td>
</tr>
<tr>
<td>Lean flow rate, L/min</td>
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<td>26.9</td>
<td>32.0</td>
<td>27.0</td>
<td>31.3</td>
</tr>
<tr>
<td>Lean MEA conc., wt.%</td>
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<td>25.5</td>
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<tr>
<td>Lean CO₂ loading, mol/mol</td>
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<td>0.314</td>
<td>0.294</td>
<td>0.284</td>
<td>0.280</td>
</tr>
<tr>
<td>Inlet flue gas flow rate, kg/h</td>
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<td>482.8</td>
<td>491.1</td>
<td>488.6</td>
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</tr>
<tr>
<td>Stripper top pressure, kPa</td>
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<td>189.2</td>
<td>180.4</td>
<td>177.5</td>
<td>189.9</td>
</tr>
<tr>
<td>Results comparison</td>
<td>Exp.</td>
<td>Sim.</td>
<td>Exp.</td>
<td>Sim.</td>
<td>Exp.</td>
</tr>
<tr>
<td>CO₂ absorption rate, kg/h</td>
<td>73.5</td>
<td>71.32</td>
<td>74.2</td>
<td>72.99</td>
<td>74.2</td>
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<tr>
<td>Reboiler temperature, °C</td>
<td>116.9</td>
<td>118.5</td>
<td>117.2</td>
<td>120.3</td>
<td>116.4</td>
</tr>
</tbody>
</table>
Specific reboiler duty, CO₂ MJ/kg  4.48  4.61  4.33  4.37  4.45  4.41  4.35  4.41  4.50  4.43
CO₂ product purity, vol%  97.7  97.74  97.6  97.27  97.7  97.76  97.8  97.96  97.7  97.73
CO₂ capture rate, %  82.89  82.57  76.20  75.95  82.28  82.35  82.63  82.79  83.95  83.99

A2: Validation of the dynamic model in Aspen HYSYS

The dynamic model is first used to do steady-state simulations without changing the input parameters. The results are compared with the steady-state results from Harun et al. (Harun, 2012). As shown in Table A2, the results obtained by the dynamic model are in good agreement with the steady-state results.

Table A2. Comparison of the results of the dynamic model and the steady state model at base case conditions.

<table>
<thead>
<tr>
<th>Lean stream</th>
<th>MEA</th>
<th>Stack gas stream</th>
<th>Rich MEA stream</th>
<th>CO₂ outlet from stripper</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (K)</td>
<td>314</td>
<td>314</td>
<td>319.8</td>
<td>333.25</td>
</tr>
<tr>
<td>Total molar flowrate (mol/s)</td>
<td>31.95</td>
<td>30.36</td>
<td>3.58</td>
<td>3.86</td>
</tr>
<tr>
<td>Mole fraction</td>
<td>CO₂</td>
<td>0.0305</td>
<td>0.0307</td>
<td>0.0083</td>
</tr>
<tr>
<td></td>
<td>H₂O</td>
<td>0.8646</td>
<td>0.8605</td>
<td>0.0944</td>
</tr>
<tr>
<td></td>
<td>MEA</td>
<td>0.1049</td>
<td>0.1098</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>N₂</td>
<td>0</td>
<td>0</td>
<td>0.8972</td>
</tr>
</tbody>
</table>

¹ Sim. means the dynamic model results. ² Ref. means the steady state results from the reference of Harun.

In addition to the steady state validation, the dynamic model is also validated with the transient data (Harun, 2012). With the input of change of FG flowrate shown in Figure A1, Figure A2 offers the validation, including the temperature profile in the absorber, CO₂ removal rate in the absorber, and energy penalty. For the absorber (Figure A2 (a)), the maximum temperature deviation is 5.9K, and the average deviation is 2.4K. For the stripper (Figure A2 (b-c)), with the increase of FG flowrate, the CO₂ removal rate decreases because the solvent is not increased to absorb more CO₂. The maximum deviation on the CO₂ removal rate is 1.6%. The energy penalty from Harun et al. (Harun et al., 2012) is calculated based on the amount of removed CO₂ in the absorber. The maximum deviation of the energy penalty is 1.6%.
Figure A1. The changes in FG flowrate (Harun et al., 2012).

(a) Temperature profile in the absorber.

(b) CO₂ removal rate.

(c) Energy penalty.

Figure A2. Validation of the Dw/oC.
A3: Methods of the model scale-up

The model is scaled up after validation, and it mainly includes the estimation of lean solution flowrate and the diameter and height of the absorber and stripper. As shown in Equation A-1, the lean solution flowrate required to capture 90% of the CO2 in the FG is estimated based on the absorption capacity of 0.2 mol CO2/mol MEA (0.287 of lean loading), lean solution MEA concentration of 30 wt%, average CO2 mass fraction of 0.186 and the largest FG mass flowrate of 100 kg/s from the w-CHP plant.

\[ L_{\text{lean}} = \frac{G_x\varphi_{\text{CO}_2}}{100z(\alpha_{\text{rich}} - \alpha_{\text{lean}})} \left[ \frac{M_{\text{MEA}}}{44.009} \left( 1 + \frac{1 - \omega_{\text{MEA}}}{\omega_{\text{MEA}}} \right) + \alpha_{\text{lean}} \right] \]  
\[ \text{(A-1)} \]

where \( L_{\text{lean}} \) means lean solution mass flowrate, kg/s; \( G \) means FG mass flowrate, kg/s; \( x_{\text{CO}_2} \) means CO2 mass fraction, %; \( \varphi_{\text{CO}_2} \) means CO2 capture rate, %; \( M_{\text{MEA}} \) means molar mass of MEA, kg/kmol; \( \omega_{\text{MEA}} \) means MEA concentration, wt%; \( \alpha_{\text{rich}} \) and \( \alpha_{\text{lean}} \) means rich loading and lean loading, mol CO2/mol MEA.

The column diameters are determined based on two criteria: (i) the maximum pressure drop that can be tolerated (flooding pressure drop) and (ii) the approach to maximum operational capacity (MOC:70%). Therefore, as shown in Equations A-2 and A-3, with a given FG flowrate and lean solution flowrate, the flooding velocity is first correlated with the flooding pressure drop and fluids’ thermophysical properties. Then, the column diameter is thus calculated.

\[ V_{G,fl} = 0.3048 \left[ \left( \frac{\rho_G}{\rho_{L}-\rho_G} \right)^{-0.5} \nu^{-0.05} F_p^{-0.5} A \left( \log \left( \frac{L}{G} \sqrt{\rho_G} \right) \right)^2 + B \left( \log \left( \frac{L}{G} \sqrt{\rho_L} \right) \right) + C \right] \]  
\[ \text{(A-2)} \]

\[ D = \frac{4G}{\pi V_{G,fl} \rho_G}, \quad V_G = 0.7V_{G,fl} \]  
\[ \text{(A-3)} \]

where \( V_{G,fl} \) means flooding velocity, m/s; \( \rho_G \) and \( \rho_L \) means gas density and liquid density, kg/m³; \( \nu \) means kinematic viscosity, cst; \( F_p \) means packing factor of column (IMTP 38 with packing factor of 78.7 m⁻¹); \( A, B \) and \( C \) are parameters correlated with packing factor (details can be found in (Otitoju et al., 2020)); \( V_G \) means superficial gas velocity, m/s; and \( D \) means the diameter, m.

The height of the absorber is calculated by the height of the transfer unit (HTU) method, as shown in Equation A-4. The height of the stripper is determined through a sensitivity analysis of reboiler duty with the change in the number of stages. Starting with a generic total stage number of 5, the number of stages in the stripper is continuously increased by 1 until a certain point where a further increase has a negligible effect on the reboiler duty.

\[ H = N_{OG} \times H_{OG}, \quad N_{OG} = \ln \frac{\gamma_{\text{CO}_2,\text{in}}}{\gamma_{\text{CO}_2,\text{out}}}, \quad H_{OG} = \frac{G_i}{K_G \ast \alpha \ast P} \]  
\[ \text{(A-4)} \]
where $H$ means the height, m; $N_{OG}$ means the overall number of the transfer unit; $H_{OG}$ means the height of the transfer unit, m; $y_{CO_2, in}$ and $y_{CO_2, out}$ means the mole fraction of CO$_2$ in the inlet gas and outlet gas; $G_i$ means the gas molar flowrate per cross-sectional area, kmol/m$^2$s; $K_G$ means the overall gas-phase mass transfer coefficient; kmol/m$^3$s; $a$ means specific surface area of packing, m$^2$/m$^3$; and $P$ means Pressure, bar.