Nonlinear Model Predictive Control of Post-combustion CO$_2$ Capture Process for Flexible Operation

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Abstract: A large amount of energy requirement for solvent regeneration is a major barrier to the widespread adoption of amine-based post-combustion CO$_2$ capture (PCC). Flexible operation is one of the ways to lower the energy penalty by responding to changes in economic factors like the energy price. However, for effective implementation of flexible operation strategies, it is necessary to identify the most economic operating condition under various potential scenarios and to establish an appropriate control strategy to operate the process. As flexible operation will inherently involve a large operating envelope, we investigate the use of nonlinear model predictive control (NMPC) technology. To circumvent the problem of solving a large-scale nonlinear programming problem online, a simpler NARX model is identified and used. With the NARX model, an offset-free NMPC is designed and simulated under various dynamic scenarios. The developed NARX-based NMPC shows satisfactory control performance, stabilizing the CO$_2$ capture rate faster than LMPC by 60-100 min.

Keywords: Post-combustion CO$_2$ capture, Dynamic simulation, System identification, Nonlinear model predictive control

1. INTRODUCTION

The interest in reducing global CO$_2$ emissions to fight climate change has brought the spotlight onto carbon capture, utilization and storage (CCUS) technologies. Complete replacement of fossil energy with renewable energy will take several decades, and CCUS technology is expected to play an important bridge role in the meantime. Carbon capture being the first step of CCUS has seen many research efforts and commercial-scale capture plants have already appeared around the world. Among various carbon capture technologies, the amine-based post-combustion carbon capture (PCC) process is the most mature technology and has been employed in capturing CO$_2$ gases emitted from power plants, one of the largest sources of CO$_2$ emission. Recent research efforts have resulted in energy and cost savings through the development of new absorbents (Muchan et al., 2017; Zhang et al., 2019), process integration (Jung et al., 2015; Oh et al., 2020) and process parameter optimizations (Agbondgae et al., 2014; Li et al., 2016). Nevertheless, the PCC process still consumes a large amount of regeneration energy to strip out the CO$_2$ from the absorbent which must be recycled for more use.

Another important aspect of the PCC process with direct economic ramifications is its operational flexibility. The input stream of the PCC process is often the flue gas from a power plant. Meanwhile, the power plant is operated to match the power generation load with the electricity demand which varies significantly at various time scales (e.g., hourly, daily and seasonally). When the demand is high, more fuels are combusted to increase the power generation, which causes an increased amount of flue gas that the PCC process should treat, and vice versa. In some cases, the opposite may happen if steam used to run the regeneration process is drawn from the power plant itself. In that case, higher power plant loads will leave less steam for the PCC process, which must respond by lowering the capture rate. Considering the wide operational range of many modern power plants, it is imperative for a power-plant-integrated PCC process to be able to absorb load changes through a flexible operation.

The improved operational flexibility also can play a major role in reducing the operating cost (Patron & Sandoval, 2021). The flexible operation strategy can be devised such that the operating condition is optimized for changing economic parameters like the energy price. It is estimated that flexible operation of the PCC process can increase the profitability by up to 16%, compared to operating at a fixed condition (Mac Dowell and Shah et al., 2015).

One of the challenges for adopting the flexible operation strategy is the complex dynamics of the PCC process. The dynamic response of the PCC process can be very slow, with a typical process requiring hours to reach a new steady-state after a change (Wu et al., 2018). From a control perspective, the process shows strong couplings among the multiple manipulated variables (MV) and controlled variables (CV) (Flo et al., 2016). The nonlinearity of the dynamics over a typical range of flexible operations can be quite strong (Jung et al., 2020). Given such characteristics of the PCC process, many studies have focused on the application of advanced control strategies (Bui et al., 2014; Wu et al., 2020).

A model-based control like model predictive control (MPC) is a good solution for handling such complex multivariable nonlinear dynamics. When a linear MPC (LMPC) is applied to the PCC process, superior control performance and satisfaction of constraints were found to be achieved compared to the traditional proportional-integral-differential (PID) controllers (Salvinder et al., 2019; Wu et al., 2020). However,
the use of a linear model can cause the control performance to degrade when the process is operated flexibly over a wide operating envelope (Jung et al., 2020). Nonlinear MPC (NMPC) may be a more attractive method for the PCC process to be operated flexibly (He et al., 2018). The extended operation window and fast closed-loop dynamics would serve to enhance the benefits of adopting a flexible operation strategy.

This study aims to show how an NMPC controller may be constructed for the PCC process which is operated over a wide operating range. The study covers from identifying a nonlinear black-box model to designing the offset-free MPC and analyzing its closed-loop performance under some set-point tracking scenarios.

2. SYSTEM DESCRIPTION

2.1 Target system

A monoethanolamine (MEA)-based PCC process, which treats a flue gas stream from a 350MWe scale natural gas combined cycle (NGCC) power plant, is a target system in this study. A dynamic process model for a typical flowsheet of the PCC process, which includes an absorber, a stripper and a heat exchanger, is implemented in the commercial software package of gPROMS (Figure 1).

![Flowsheet of a typical PCC process](image)

In Figure 1, the flue gas emitted from the NGCC power plant is introduced as an input stream at the bottom of the absorber. 30wt% MEA solvent stored in a buffer tank is introduced to the top of the absorber as a counter-current flow and selectively absorbs the CO₂ contained in the flue gas through exothermic chemical reactions. Cleaned gas goes out from the top of the absorber, while the CO₂-rich MEA solvent is heated in the heat exchanger. The CO₂ in the heated solvent is desorbed in the stripper by additional regeneration heat supplied from the reboiler. The regenerated CO₂ lean solvent is then recycled and stored in the buffer tank, after passing through the heat exchanger for heat recovery.

For the dynamic process model, a rigorous rate-based column model validated in Jung et al. (2020) is used for the absorber and the stripper, while a logarithm mean temperature difference (LMTD) model is used for the heat exchanger model. The configuration of each process unit is designed based on the nominal operation conditions, which can be found in Table 1 along with the corresponding unit configurations.

| Type               | Variable            | Value | Units |
|--------------------|---------------------|-------|-------|
| Inlet stream       | Flue gas flowrate   | 659   | kg/s  |
|                    | CO₂ in flue gas     | 4.2   | mol%  |
|                    | H₂O in flue gas     | 6.6   | mol%  |

| Process unit config | Absorber         | Value | Units |
|--------------------|------------------|-------|-------|
| - Height            | 22               | m     |
| - Diameter          | 14               | m     |
| - Packing type      | Mellapak 250Y    |       |

| Nominal condition | CO₂ capture rate  | 90    | %     |
|                  | Stripper pressure | 180   | kPa    |
|                  | Temperature approach | 10     | °C    |
|                  | Solvent flowrate  | 765   | kg/s  |
|                  | Reboiler temperature | 118.3  | °C    |
|                  | Regeneration duty | 157   | MW    |
|                  | Lean loading      | 0.236 | mol/mol |
|                  | Rich loading      | 0.476 | mol/mol |
|                  | CO₂ purity        | 96    | mol%  |
|                  | Specific regeneration duty | 4.0 | MJ/kgCO₂ |

In Table 1, the CO₂ capture rate ($\eta_{CO₂}$) and specific regeneration duty (SRD) are defined as (1) and (2), respectively.

$$\eta_{CO₂} = 1 - \frac{F_{CO₂, outlet}}{F_{CO₂, inlet}} \quad (1)$$

$$\text{(SRD)} = \frac{Q_{reg}}{F_{CO₂, inlet} - F_{CO₂, outlet}} \quad (2)$$

where $F_{CO₂, inlet}$ and $F_{CO₂, outlet}$ represent the inlet and outlet CO₂ flowrate in the absorber unit respectively. $Q_{reg}$ notes the regeneration duty in the reboiler.

2.2 Flexible operation range

To operate the PCC process flexibly in the face of process disturbances (e.g. inlet flue gas conditions) and/or varying external economic parameters (e.g. CO₂ penalty cost, energy price), an operating range must first be set within which the process with the controller can maintain stable operation.
Table 2 gives the target operating range of the PCC process that the controllers should work.

The flue gas flowrate emitted from a power plant is approximately proportional to the plant load (Mac Dowell and Shah, 2013). A typical NGCC power plant can reduce the operation load by about 50% of its maximum load (Henderson, 2014). Since operation beyond the maximum load of the power plant is not considered, 50-100% of the nominal flue gas flowrate falls within the targeted operating range. The possible operating ranges of the solvent flowrate and the reboiler duty are assumed to be 50-150% of their nominal values, with the resulting operable range of the CO2 capture rate being 30-99%.

| Variable              | Operating range | Units |
|-----------------------|-----------------|-------|
| Flue gas flowrate     | 330 – 659       | kg/s  |
| Solvent flow rate     | 382 – 1201      | kg/s  |
| Reboiler duty         | 79 – 235        | MW    |
| CO2 capture rate      | 30 - 99         | %     |
| Reboiler temperature  | 377 - 395       | K     |

3. SYSTEM IDENTIFICATION

3.1 System identification with NARX

A model-based controller, e.g. MPC, requires a dynamic model to predict the future behavior of the controlled system and the optimal control actions through dynamic optimization. However, directly using the rigorous process model (used for the process simulation) in the control algorithm is very difficult as the model is too complex (108066 model equations, 520 of which are differential and 107546 are algebraic) to be used for the online optimization. Rather than solving the complex DAE system, the use of a data-driven model enables the online optimization within a limited time with a low computational load. To implement MPC, a simple data-driven model is identified using data generated by the rigorous process model. In this study, a nonlinear autoregressive with exogenous variables (NARX) model (Chen and Billings, 1989) is chosen for the model structure. The NARX model is a structure widely adopted in studies of data-driven nonlinear modeling and control applications (Chen et al., 1990). It relates the unknown mapping of \( f \), which correlates the past variables to the future variables, can be parameterized in various forms such as linear, polynomial, and so on. In our case, given the complexity of the dynamics, a multi-layer perceptron (MLP) is used to represent the mapping function of \( f \).

\[
y_{t+1} = f(y_t, y_{t-1}, y_{t-2}, \ldots, y_{t-n_y}, u_t, u_{t-1}, u_{t-2}, \ldots, u_{t-n_u}) + \epsilon_t
\]

where \( y_t \) and \( u_t \) represents the output and input variables at time \( t \) respectively, and \( n_d \) represents the maximum lags included. The unknown mapping of \( f \), which correlates the past variables to the future variables, can be parameterized in various forms such as linear, polynomial, and so on. In our case, given the complexity of the dynamics, a multi-layer perceptron (MLP) is used to represent the mapping function of \( f \).

The rigorous process model is first simulated by varying the input variables to generate a learning data set for the NARX model. The input variable set \( u_t \) includes the inlet flue gas flowrate \( (F_{\text{flue}}) \), the solvent flowrate \( (F_{\text{solv}}) \) and the regeneration duty \( (Q_{\text{reg}}) \). The three variables are randomly perturbed with step or ramp signals within the range of Table 2 at every 100 min for 4000 min. The output variable set \( y_t \) which includes the outlet CO2 flowrate \( (F_{\text{CO2, outlet}}) \) and the reboiler temperature \( (T_{\text{reb}}) \) are obtained from the simulation of the rigorous model. During the simulation, all of the variables contains ±5% white noise. The dynamics of the PCC process are quite slow, so the sampling time is set to be 1 min. Although one can directly use the CO2 capture rate as the output variable of the NARX model, the choice increases the nonlinearity of the mapping function \( f \) as shown in (1). Since \( F_{\text{CO2, inlet}} \) used in the calculation of \( \eta_{\text{CO2}} \) can be obtained from \( F_{\text{flue}}, F_{\text{CO2, outlet}} \) is chosen as the output variable to alleviate the model nonlinearity. The time lag \( n_y \) of the NARX model is set at 3. The NARX model to be identified for the PCC process is a 3 by 2 multi-input, multi-output (MIMO) system. However, the overall NARX model is formulated as an ensemble of the multi-input, single-output (MISO) NARX models to avoid the loss-of-identifiability problem which is known to exist even for linear multi-output time-series models.

3.2 Multi-step ahead prediction

The key of the NARX model is that it finds the correlations between past or present values of the variables and future variables. Assuming a time-invariant dynamic system, the NARX model can be used to construct multi-step predictions of the outputs by iteratively applying the one-step prediction model. However, the accuracy of the multi-step prediction may deteriorate as the one-step-ahead prediction error is accumulated in the prediction. For this reason, we need to examine the accuracy of the multi-step ahead predictions before applying the identified NARX model to the controller.

![Model prediction and validation dataset](image)
The 20-step ahead prediction results are compared to the validation datasets. The model predicts the dynamic characteristics or trends well without noticeable outliers (Figure 2) and the identified NARX model shows good agreements with the validation data. The 20-step prediction of the NARX model has R² values of 0.995 and 0.991 for outlet CO₂ flowrate and reboiler temperature, respectively (Figure 3). The identified NARX model is judged to have sufficiently accurate multi-step prediction capability to be used in the MPC algorithm, replacing the original complex dynamic process model.

![Graph](image)

**Figure 3** Comparison of normalized model prediction and validation dataset (20-step ahead prediction); a) outlet CO₂ flowrate; b) reboiler temperature

### 4. SET-POINT TRACKING CONTROL

#### 4.1 Control problem

Our control problem is first defined in this section. For the flexible operation of the PCC process, the amount of CO₂ captured in the absorber should track the desired value, even with large changes in the inlet flue gas flowrate due to power plant load changes. At the same time, the reboiler temperature must be strictly bound at a designed value, since it is the main process variable that determines the regeneration quality of the recycled lean solvent. Hence, the CO₂ capture rate and the reboiler temperature are the controlled variables. To control these variables, the recirculated lean solvent flowrate and the regeneration heat are used as MVs. The inlet flue gas flowrate is assumed to be a measured disturbance, which provides feedforward information to the controller. Table 4 shows a clear definition of the set-point tracking control problem.

Those variables that are related to process unit inventories (e.g. tank level, pressure) are assumed to be controlled tightly with simple PID controllers to simplify the control problem.

| Type                | Variable   |
|---------------------|------------|
| Control variable    | \( n_{CO2}, T_{reb} \) |
| Manipulated variable| \( F_{sol}, Q_{reg} \) |
| Measured disturbance| \( F_{flue} \) |

#### 4.2 Nonlinear model predictive control

Using the identified NARX model as a predictive model directly for the MPC calculation can lead to undesirable results. To design a set-point tracking controller appropriately, it is necessary to consider possible errors between the model and the actual process, which can come from a model-plant mismatch or unmeasured disturbances. If one designs NMPC without taking these errors into account, the controller may not provide correct feedback actions and offsets may result in the closed-loop dynamics (Pannochia et al., 2015). To design an offset-free MPC, model augmentation with a simple output disturbance term can be introduced as shown in (4).

\[
y_p(k+i|k) = y_m(k+i|k) + d(k)
\]

\[
y_m(k+i|k) = f(y_m(k+i-1|k), \ldots, y_m(k+i-n_d|k), u(k+i-1), \ldots, u(k+i-n_d))
\]

where \( y_p \) and \( y_m \) represents the predicted and NARX model outputs respectively. The output disturbance at time \( k \), \( d(k) \) can be estimated as follow:

\[
d(k) = d(k-1) + L_d(y(k) - y_m(k|k-1))
\]

where \( y \) represents the process measurements. \( L_d \) is a parameter for the correction rate, which is given as 0.1 in this study. By introducing the disturbance term and updating it every sampling time, bias in the model prediction along the predictive horizon can be corrected using the measurement at time \( k \). Although this approach cannot guarantee the removal of offsets in all cases, it is widely used in practice due to its clarity and simplicity (Tian et al., 2014).

With the modified predictive model, the NMPC problem solved at every sampling time is defined as the constrained nonlinear optimization problem, where the future control actions are determined by minimizing the following objective function:

\[
\min_u \quad J = \sum_{i=1}^{p} (y_p(k+i|k) - y_r)^T Q(y_p(k+i|k) - y_r) + \sum_{i=1}^{m} \Delta u(k+i|k) R \Delta u(k+i|k)
\]

subject to the following constraints:

\[
y_{min} \leq y_p(k+i|k) \leq y_{max}
\]

\[
u_{min} \leq u(k+i|k) \leq u_{max}
\]

\[
\Delta u_{min} \leq \Delta u(k+i|k) \leq \Delta u_{max}
\]

where \( p \) and \( m \) are the sizes of the prediction and control horizon, \( y_r \) is the set-point vector for the controlled variables, and \( Q \) and \( R \) are the weighting matrices on the output errors and the input movements. The control input movement, \( \Delta u(k+i|k) = u(k+i|k) - u(k+i-1|k) \) is considered only within the control horizon of \( m \), and no further input change is assumed beyond that point. During time integration of the predictive model, the measured disturbance is considered to be constant. In the MPC problem formulation, the sampling time is set to be 1 min, given the time scale of the dynamics of the PCC process. The prediction and control horizons are set to 20 and 3 respectively. The weighting matrices \( Q \) and \( R \) are set to
be \texttt{diag([1, 1])} and \texttt{diag([5, 5])} respectively. They are not optimized through search; they are determined based on intuition and heuristics. The optimization problem is solved with the interior point method, which is a popular optimization algorithm for a convex problem. Although there is no guarantee that the optimization problem of (6) is convex, the calculation time is limited.

4.3 Simulation study

Since the rigorous plant model is implemented in gPROMS, while NMPC is formulated and computed in MATLAB, we have to weave the two different simulation environments for the closed-loop simulation study. gPROMS sends the CV values to MATLAB, and the NMPC algorithm in MATLAB calculates the next time MVs based on the sent CVs. These actions are repeated at every sampling time to conduct a closed-loop simulation.

To compare the performance of the developed NMPC controller, an LMPC controller was also designed and simulated. A linear dynamic model in the LMPC controller was obtained by linearizing the process model at the nominal operating condition. To estimate the state of the system without offsets, both input and output disturbance models were used and the Kalman filter was designed for state estimation. The main controller parameters such as prediction horizon or weights were set to be the same as those in the NMPC controller.

A dynamic scenario was considered to verify closed-loop control performance of the NMPC controller. The scenario includes both a load disturbance and set-point change. A disturbance or set-point change occurs every 200 minutes, and the total scenario lasts 1200 minutes. The disturbance is introduced to the flue gas flowrate, which decreases to 80% at 200 minutes and returns to the nominal value at 800 minutes. This disturbance instantiates a case where power generation load of the NGCC plant varies due to time-varying electricity demand. For a set-point change, the target CO2 capture rate changes from 90% to 60%, 95% and back to 90%. The large-size change in the CO2 capture rate reflects a flexibly operated PCC process that responds actively to changes in energy prices or steam availability.

The simulation result (Figure 4) shows that control performance of NMPC is superior to that of LMPC, especially for CO2 capture rate, which is the main controlled variable. This difference becomes noticeable at the 200 and 800 min marks when the operating condition changes significantly. The closed-loop dynamics of the CO2 capture rate show large deviations at those times when LMPC is applied. On the other hand, NMPC can quickly stabilize the process to a steady state within about 60 minutes. Due to the nature of model-based predictive control, NMPC which correctly reflects nonlinear system dynamics can regulate controlled variables more tightly than LMPC. In particular, the result shows that NMPC is advantageous to LMPC to cope with disturbances in flue gas flowrate, which typically occur in a PCC process which is energy-integrated with a power plant.

Another noteworthy observation can be made during the time period from 400 to 600 minutes when the operating condition is changed significantly from the nominal point. LMPC shows oscillating dynamics due to a large gap between the linearized predictive model and the nonlinear process dynamics. Oscillations can occur when the process gain changes significantly, and it is known that the gain values of the PCC process have large sensitivities depending on the operating conditions (Jung et al, 2020). An increased weight on the control action rate can remove the oscillation, but it can make the control performance very sluggish. In the end, using NMPC can help improve the overall closed-loop dynamics of the PCC process, stabilizing the process faster than LMPC by about 60-100 minutes.

5. CONCLUSIONS

NARX-based NMPC was implemented to control the PCC process in the face of large inlet flowrate and set-point changes. The rigorous first-principles model of the process was developed but judged to be too complex for direct use in NMPC, so a simpler NARX model was identified using data generated from the rigorous model. Using the identified model, an NMPC controller was designed and tested under certain disturbance and set-point tracking scenarios. The closed-loop performance of NMPC was compared to that of LMPC, and revealed a superior control performance. The difference in control performance was distinctive when a disturbance in flue gas flowrate occurred and when the operating condition deviated significantly from the nominal condition. Results of the simulation study indicate that nonlinear dynamics of the PCC process exposed by the extended operation window can greatly degrade performance of a linear controller, and that application of the more complex NMPC technology is warranted in order to tightly control the PCC process in the face of large operating condition changes.
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