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## Dynamic model of a post-combustion absorption unit for use in a non-linear model predictive control scheme

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### Abstract

With an increasing demand on load flexibility in power supply networks, advanced control systems for plants with carbon capture units gain in significance. Minimizing the energy demand for carbon dioxide removal under these circumstances is a major task of such a control strategy. In this work a dynamic model in Modelica of a chemical absorption process run with an aqueous monoethanolamine (MEA) is developed. Starting from a rather detailed dynamic model of the process, model reduction is performed based on physical insight. The reduced model computes distinctly faster, shows similar transient behavior and reflects trends for optimal steady-state operations reported in the literature. The model is intended to be used in the framework of JModelica.org, a platform supporting non-linear dynamic optimization.

© 2011 Published by Elsevier Ltd. Open access under [CC BY-NC-ND license](https://creativecommons.org/licenses/by-nc-nd/4.0/).Keywords: CO<sub>2</sub>, absorption, model, optimization, nonlinear model predictive control, Modelica

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### 1. Introduction

Carbon dioxide (CO<sub>2</sub>) removal from a gas mixture using aqueous amine solutions is a well established process previously mainly used for gas sweetening in refineries. Although the focus there lies primarily on the removal of hydrogen sulfide it is equally applicable to flue gas from fossil-fuel fired power plants.

Figure 1 shows a schematic of the process. The CO<sub>2</sub> from the flue gas is absorbed by the liquid solvent in the absorber column. The cleaned gas is released to the environment, while the rich solution is transported to the desorber column. Here, the CO<sub>2</sub> is released at elevated temperatures to a gas flow from the reboiler, which utilizes steam from the power generation process. The product CO<sub>2</sub> stream is then compressed and stored. The overall power plant efficiency is expected to be reduced by at least 10 %, the solvent regeneration being responsible for more than half of this [1]. Minimizing the amount of steam required in the reboiler is therefore a primary task in the optimization of this process.

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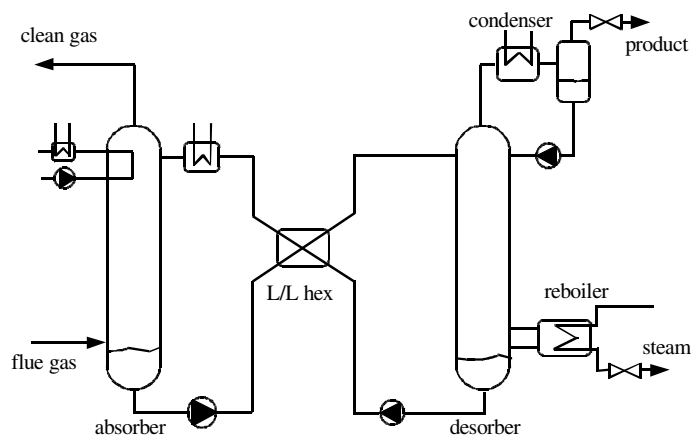


Figure 1: Schematic of an absorption/desorption process to remove carbon dioxide from power plant flue gas.

With an increasing demand on the plant's flexible operation in the face of frequent load changes dynamic simulation and optimization have become important tools to ensure an efficient incorporation of the carbon capture into the power generation. At the same time a trade-off must be found between efficiency losses and removal rate, possibly governed by economic boundary conditions.

The paper presents the preliminary results achieved within a larger project aiming at developing an optimization technology for advanced model-based control of the separation plant. The paper focuses on the modeling of the capture plant and gives also a short description of the methods and the tools that are going to be used for the optimization.

## 2. Background

### 2.1. Modeling of carbon dioxide removal with chemical absorption

System simulation models of amine scrubbing processes with different levels of detail can be found in the literature and as part of commercial toolboxes. The most rigorous models are developed for steady-state system computations with partial differential equations for mass transport along bulk flow and between the two phases, resulting in a high order system. This becomes easily too complex for dynamic system simulations, especially if parts of the power generation are supposed to be included or if used in model based control. Replacing rigorous models of multi-component mass transfer between gas and liquid with semi-empirical algebraic correlations reduces model complexity dramatically and is for example applied in [2] for an absorber description. Another model aspect with room for different levels of detail is the thermodynamic model of the liquid phase, describing the non-ideality of the electrolyte solution. Tobiesen compares in [3] a more rigorous with simpler approaches and concludes that high accuracy is rather a matter of a good data fit than model complexity.

Several studies on optimal operation of an amine-based CO<sub>2</sub> capture plant can be found in the literature. In [4] the effect of variables such as solvent circulation rate, stripper pressure or solvent temperature is investigated. The analysis is however static and considered only the variation of one parameter at a time, disregarding the multivariable and dynamic nature of the process. In [5] control strategies aiming at a fast response are developed using offline dynamic simulation of the process. In [6], both optimization and control of the plant are studied. The optimal conditions for operation are determined offline using static models and a suitable control structure to maintain the process close to optimal operation in spite of disturbances is thereafter derived using dynamic models.

### 2.2. Modeling with Modelica

The Modelica language has evolved from the simulation community, with roots in analog simulation dating back to the 1940's. The first version of Modelica was published in September 1997. The effort was targeted at creating a new general-purpose modeling language, applicable to a wide range of application domains. While several other modeling languages were available, many of those were domain-specific, which made simulation of complex heterogeneous systems difficult. Based on experiences from designing other modeling languages, the fundamental concepts of object-orientation and declarative programming were adopted. The latest version of the Modelica specification, 3.2, (see [7]) was released in 2010.

Modelica is an object-oriented language in that it uses objects, classes, and inheritance to define and specialize models of physical entities. A fundamental difference from an ordinary programming language, like Java, is the way

behavior is defined: In Modelica, there is no concept of run-time state, dynamic object allocation, or method dispatching. What a Modelica program does is to define a number of statically allocated objects, called “components” in Modelica terminology. The behavior of the individual components is defined using differential and algebraic equations that typically capture laws of nature. Classes and multiple inheritance are used for abstracting and specializing the components and their behavior.

### 2.3. Model Predictive Control

Model Predictive Control (MPC) has established itself as one of the most commonly used advanced control strategies in the process industry [8]. At the heart of an MPC controller is an algorithm that computes the solution to an open loop optimal control problem with a fixed time-horizon. The first sample of the optimal control profiles is then applied to the plant. At the next sample, the procedure is repeated, and a new optimal control problem is solved based on updated state estimates. Application of an optimization horizon that is shifted in each sample has rendered MPC to also be referred to as Receding Horizon Control.

Two major advantages of using MPC are that multiple input multiple output (MIMO) systems are handled consistently and that constraints acting on controls and measured variable can be taken into account explicitly. MIMO systems are common in the process industry, where typically several control variables are available as well as several, sometimes contradictory, control objectives need to be fulfilled. The task of the control designer is then to tune the weighting functions of the optimal control problem to match the objectives, which may be formulated as economic measures or as to minimize operating energy or raw material consumption. In addition, constraints are frequently occurring, e.g., limited range of actuators, pressures that need to be kept within safety limits and tanks that should not overflow. The ability to model such constraints explicitly in the formulation of an MPC controller is therefore a key strength.

The key to executing MPC controllers is the solution of an optimal control problem that in turn is based on a dynamic model of the plant to be controlled. This task is often very computationally demanding, especially if the system is large and exhibits non-linear behavior. In fact, this is one of the reasons why MPC has had particularly large success in the process control community where plants typically have slow dynamics allowing for long computation times. There are however algorithms for reducing the computational delay, see e.g. [9] and [10]. The area of devising fast MPC algorithms remains an active area of research. But even so, it is important to derive simple, yet sufficiently accurate models.

Even though MPC was initially developed within the industrial community, a large body of theory that has been developed in the academic community during the last decades supports the method. For example, results for analyzing and for guaranteeing stability are available [11]. See also [12] for a recent monograph offering an extensive treatment of the subject.

### 2.4. JModelica.org and Optimica

The software platform intended to be used in this work to solve dynamic optimization problems is JModelica.org [13]. The platform is based on the Modelica language and therefore fits well with the approach taken in this work. The main reason for choosing this platform is, however, that it offers strong support for solution of dynamic optimization problems, which is a key component of executing MPC controllers, as discussed above.

In particular, JModelica.org supports an extension of Modelica entitled Optimica [14], which allows dynamic optimization problems to be formulated based on Modelica models, in a high-level language. Optimica enables the user to express cost functions, constraints, and what to optimize in a description format that is complimentary to Modelica’s support for dynamic modeling. In the context of this work, this feature enables shorter design cycles since more effort can be put into formulation of optimization problems rather than encoding them in a specialized format for a particular optimization algorithm.

JModelica.org implements a direct collocation method [15], for solving large-scale dynamic optimization problems. The algorithm employs full discretization of both the state and control spaces, which results in a very large, but sparse Non-Linear Program (NLP). Despite the size of the NLP, efficient algorithms for solving such problems exist; in this work, the algorithm IPOPT [16] is being used. It remains to investigate, whether this method is the most appropriate for this particular application with the JModelica.org platform.

In terms of user interaction, JModelica.org offers a Python [17] interface. Using Python, Modelica and Optimica models can be compiled into executable optimization programs, optimization algorithms can be invoked and the results can be loaded. Python also comes with packages for numerical computations and visualization, which makes it a suitable environment for scientific computations. It can be noted that the capabilities of Python go beyond scripting and atomization in that full-fledged applications with customized user interfaces can be created.

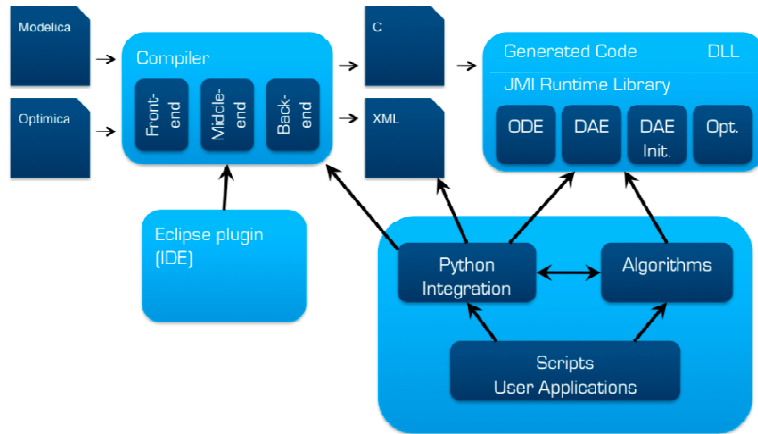


Figure 2: Schematic of the JModelica.org architecture

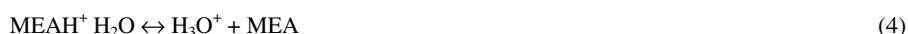
### 3. Dynamic model

The development of the carbon capture process model to be used in the optimization tool is a step-wise iteration process to reach a level of detail that is a trade-off between model accuracy and complexity with the specific application and tool capacity in mind. Since this work is still ongoing a snapshot of this process will be presented here.

The starting point is a dynamic model of an absorption unit developed and simulated in the Modelica environment Dymola. The model consists of the main components absorber, stripper, reboiler and internal heat exchanger as well as auxiliary equipment such as pumps, valves, flow resistances, cooled vessels, sensors and reservoirs, as sketched in Figure 1. The solvent is an aqueous MEA solution. The reboiler is treated as an equilibrium flash stage, the column models consist of rate driven mass transfer between gas and liquid bulk flows in packed sections. The balance equations of mass and energy conservation are discretized along the bulk flow directions. The following assumptions apply:

- all reactions occur in the liquid phase and are assumed to be in equilibrium
- the flue gas into the absorber contains only carbon dioxide, water, oxygen and nitrogen
- MEA is non-volatile and not present in the gas phase
- the total amount of liquid in the column is defined as the packing hold-up and the sump liquid volume
- the liquid in the column sumps and other large volumes is assumed to be ideally mixed
- mass and heat transfer between liquid and gas phase is restricted to packed section
- negligible temperature difference between liquid bulk and interface to gas phase
- no storage of mass and energy in the contact film
- perfect gas law applies in the gas phase
- all liquid from the packing bottom in the stripper is fed to the reboiler with a constant liquid level
- constant target packing hold-up

The liquid phase model consists of 9 species, the molecules water (H<sub>2</sub>O), carbon dioxide (CO<sub>2</sub>) and monoethanolamine (MEA) and 6 ions that are formed according to the following reactions:



A modified Pitzer equation [19] is used to compute the activity coefficients for the individual species. Full speciation is computed in the liquid bulk. The molecular carbon dioxide concentration  $c_{\text{CO}_2,b}$  is then used to compute mass transfer between bulk and interface (if).

$$\dot{n}_{iL} = A_{if} k_{iL} E (c_{i,b} - c_{i,if}) \quad i = \text{CO}_2 \quad (6)$$

$$\dot{n}_{iV} = \frac{A_{if} k_{iV} (p_{i,b} - p_{i,if})}{RT} \quad i = \text{CO}_2, \text{H}_2\text{O} \quad (7)$$

where  $\dot{n}_{iL}$  and  $\dot{n}_{iV}$  denote the molar flows in the liquid and the vapor phase, respectively.  $A_{if}$  is the contact area,  $E$  is an enhancement factor describing the impact of chemical reactions on the concentration profile near the interface.  $k$  is a mass transfer coefficient,  $c_{i,if}$  and  $c_{i,b}$  are molar concentrations at the interface and in the liquid bulk, respectively and  $p_{i,if}$  and  $p_{i,b}$  are correspondingly partial pressures of the considered species in the gas phase.  $R$  and  $T$  are the ideal gas constant and bulk phase temperature, respectively. Phase equilibrium at the gas-liquid interface for both, water and carbon dioxide, is computed as follows, assuming the pointing-factors and gas phase fugacity coefficients being equal to one.

$$y_i p = \gamma_i x_i H e_i \quad i = \text{CO}_2 \quad (8)$$

$$y_i p = \gamma_i x_i p_{i,sat}(T) \quad i = \text{H}_2\text{O} \quad (9)$$

with the mole fractions in gas and liquid phase  $y_i$  and  $x_i$ , the Henry-coefficient for dissolution of  $\text{CO}_2$  in water  $He$ , the vapor pressure of water  $p_{i,sat}$  and the system pressure  $p$ .

The enthalpy of reaction released during absorption and required to regenerate the solvent in the stripper is received from the van't Hoff equation

$$\frac{d \ln K}{dT} = \frac{\Delta H_r}{RT^2} \quad (10)$$

with the equilibrium constant  $K$  and the enthalpy of reaction  $\Delta H_r$ . The enthalpy of physical solution is computed accordingly using the temperature dependency of the Henry-coefficient [18]. Properties and correlations from the literature used in these models are listed in the table below.

Table 1: References for physical properties used in the model

Property	Symbol	Used in reduced model	Reference
Equilibrium constants	$K_i$	indirectly	Collected in [19]
Henry-coefficient	$He_{\text{co}_2}$	yes	[19]
Activity coefficients, liquid phase	$\gamma_i$	indirectly	[19]
Mass transfer coefficients	$k_{iL}, k_{iV}$	no	[20]
Diffusivities liquid phase	$D_{iL}$	no	[21] + Stokes – Einstein relation
Diffusivities gas phase	$D_{iV}$	no	Fuller's eq.in [22]
Densities and viscosities, liquid	$\rho, \mu$	yes	[23]
Enhancement factor	$E$	no	[2]

The complexity of a dynamic system model influences the computational effort to solve this problem in several ways. The resulting system of DAEs (Differential Algebraic Equations) of index 1 consists of differentiated (also referred to as dynamic states) and algebraic variables interrelated by non-linear dependencies, which need to be solved iteratively at each time step. An example for a rather computational extensive non-linear system of equations is the liquid phase speciation if chemical equilibrium is assumed for all reactions. It then also requires a good choice of iteration variables since species concentrations vary by many orders of magnitude.

Online optimization as it is used in MPC implicates tighter limitations on the model size than pure dynamic simulation or even offline optimization would do. The solution of the optimization problem for a finite horizon needs to

be found between two sampling instants and therefore demands a relatively low computational effort. But also the available memory to store result points for all model variables for each time step within the finite horizon limits the allowed number of algebraic and differentiated variables. However, exact numbers are hard to define in advance. At the same time accuracy demands are not as high as the model is updated with measurement values at each sample step.

The following measures are taken in order to reduce the model:

1. Chemical equilibrium computation (and ion speciation) was replaced by a spline approximation of the molecular CO<sub>2</sub> concentration in the liquid phase as a function of temperature and solvent loading with CO<sub>2</sub>. The mass fraction of MEA in the unloaded solution is kept constant at 30% for this function.
2. Enthalpy of absorption/desorption is replaced by a function of temperature but constant over the entire range of solvent loading.
3. Mass transfer coefficients including enhancement by chemical reactions are no longer computed from physical medium properties, but become constant tuning parameters.
4. Reduction of the number of volumes in bulk flow direction to an acceptable minimum (iterative, dependent on application)

The number of dynamic state variables is only influenced by the discretization, while a distinct reduction of algebraic variables can be achieved by bringing the number of species in the liquid phase from 9 including ions down to 3. Both models were simulated with a step-wise increase of the flue-gas inlet mass flow rate. The inlet CO<sub>2</sub> concentration was 13 Vol-%. Mellapak250Y was used in both column models with active packing heights of 8 m and a diameter of 1 m. In Figure 3 the removal rate is plotted versus simulated time for both cases. Figure 4 compares the CPU times for both models on a standard notebook.

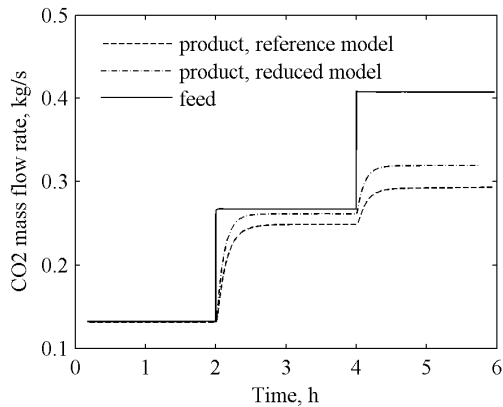


Figure 3: CO<sub>2</sub> removal from flue gas feed, reference and reduced model, constant reboiler duty

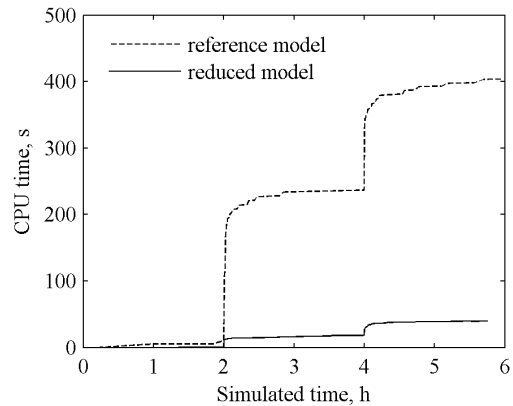


Figure 4: Used CPU time vs. simulation time for both model variants and boundary conditions as in figure 3

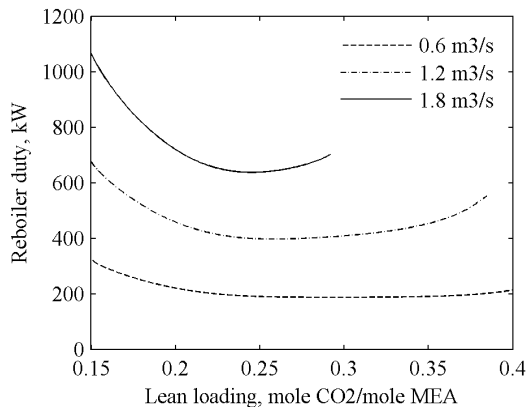


Figure 5: Reboiler duty with respect to lean loading at different flue gas flow rates and constant removal rate of 90%

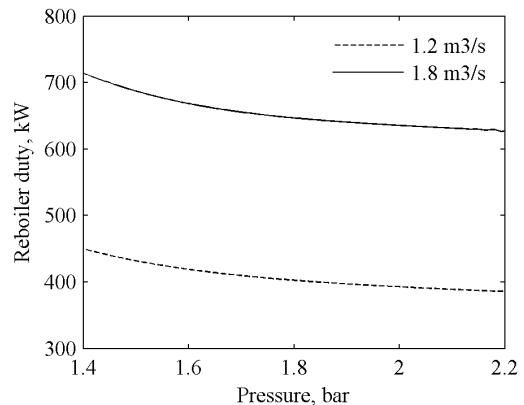


Figure 6: Reboiler duty vs. stripper top pressure at a constant removal rate of 90% and 0.25 mol/mol lean loading

The constant mass and heat transfer coefficients in the reduced model have not been tuned specifically to the reference model yet, deviations can be found in the removal rate simulated with the two models (Figure 3). However, the transient response of the carbon dioxide product flow rate reveals similar dynamic behaviour. The CPU time needed to perform the simulation decreases by a factor of around 10. Investigations carried out on steady-state process optimization ([4], [24]) propose the lean loading as a good candidate for optimizing the steam consumption. Figure 5 shows simulation results of the reduced model. The reboiler duty was controlled to follow an extremely slow variation of the lean loading in the stripper bottom, to ensure a near steady-state operation, while the removal rate was kept constant at 90% by adapting the solvent recirculation rate. For three different flue gas feed rates optimal lean loadings could be observed in regions also found in the literature, more prominent ones at high throughput rates. Figure 6 shows the reboiler duty at a constant lean loading and removal, but variable stripper head pressure. Due to MEA degradation promoted by high temperatures the pressure is usually restricted to around 1.8 bar, the simulations reflect the known tendency for lower energy consumption at higher pressures.

#### 4. Formulation of the optimization problem

The objective of the optimization is to minimize the operational cost of the capture plant under both static and dynamic conditions. The cost function takes typically into account the power consumption of the pumping devices, the cost associated with CO<sub>2</sub> emission, the steam utilized in the reboiler, the fuel and the electricity prices [1].

Looking at the scheme in Figure 1 a total of 9 degrees of freedom can be identified for both control and optimization purposes: valves and pumps in gas, solvent or cooling water flows. Some variables are used directly for level control in absorber, desorber and condenser, as well as temperature constraints, thereby reducing the size of the optimization problem, but the resulting optimization problem is still a complex one with many dynamic (Figure 3) and nonlinear (Figure 5) interactions, motivating a global and multivariable approach as MPC.

As far as the optimization constraints are concerned, they are of both regulatory and operational nature. The versatile JModelica.org platform allows us to include any constraint that can be expressed in terms of process variables. A maximum CO<sub>2</sub> concentration in the flue gas or a minimal removal rate can easily be described as hard regulatory constraints in the MPC formulation. In addition to the process model equations, the following operational constraints are part of the optimization problem:

1. A maximal temperature in the stripper and reboiler (due to MEA degradation)
2. The limited capacity of the pumps
3. A maximal solvent level in absorber, desorber and condenser

Once the optimization problem is set-up, different scenarios such as load changes, variations in electricity, fuel or CO<sub>2</sub> prices will be considered to demonstrate the ability of the MPC controller to operate the plant in the most economical manner when the boundary conditions are time-varying. In a scenario where the power plant is operated in a highly dynamic mode, a complete shut-down off the capture process may be an advantageous measure under some load situations.

#### 5. Conclusions

A reduced dynamic model of a post-combustion carbon capture plant based on chemical absorption was presented. It represents a snapshot of an ongoing project where a model suitable for non-linear model predictive control within the JModelica.org platform is developed. The reduced model reveals steady-state deviations from the reference model with a close agreement of its transient characteristics, while at the same time performing 10 times faster. Model tuning against measurements or other more detailed models is expected to improve steady-state performance. Studying the lean loading in the stripper as a possible control variable, simulation results indicate local minima for the energy consumption in the reboiler. Further development will aim at iteratively finding a good agreement of model accuracy, efficiency, complexity and relevance while working with the JModelica.org platform.

The system boundary of the model presented was chosen to reflect the entire separation process. For control purposes, it could be beneficial to restrict it to parts of the system, e.g. the stripper unit with reboiler and condenser. Alternatively, it could be extended to adjoining processes if the optimal control strategy also comprises product compression and steam generation.

## 6. Acknowledgements

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