Dynamic Modeling and Validation of a Precombustion CO_2 Capture Plant for Control Design

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1. INTRODUCTION

The generation of power and heat was identified as the largest producer of CO_2 emissions with a share of more than 40% of the global CO_2 emissions in 2010.[1] With the energy demand expected to rise in the coming decades because of the rapid growth of the world population and the emergence of developing countries, nonemitting sources become essential in order to reduce the emission intensity. The International Energy Agency (IEA) estimated that carbon capture and storage (CCS) applied to fossil-fuelled power plants is a potentially beneficial technology, because it might contribute to the required reduction of CO_2 emissions for as much as 17% by year 2035.[2] This is based on the so-called 450 scenario, limiting the long-term temperature increase to 2 °C in comparison to the preindustrial level.

Among the CO_2 capture technologies, precombustion CO_2 capture applied to integrated gasification combined cycle (IGCC) power plants is a promising technical solution due to its potential for high net efficiency,[3] fuel flexibility, and low emissions of other air pollutants. The integration of the CO_2 removal unit into the very complex gasification process and combined cycle power plant leads to many technical challenges especially regarding dynamic operation. Nowadays, dynamic performance of fossil-fuelled power plants becomes increasingly important as the share of electricity produced by renewable energy sources, which is inherently unsteady, is steadily growing.[4,5] Therefore, the capture process has to be able to follow frequent and fast load changes without restraining the performance of the IGCC power plant and violating environmental requirements, which are expected to be more strict in future.

The desired dynamic performance of the capture unit can be achieved by adequate process, equipment, and control system design. The state-of-the-art approach to this type of design problem is by means of dynamic process modeling and simulation, if possible accompanied by an experimental campaign to facilitate model validation. Simulations of transient operational scenarios are indispensable to compare the performance of different process configurations, to test different control strategies, including control parameter tuning, and to perform dynamic process optimization.[6]

To investigate the transient performance of precombustion CO_2 capture units among others, a unique, fully instrumented CO_2 capture pilot plant was realized at the Buggenum IGCC power station in The Netherlands by the utility company Vattenfall.[7] Detailed dynamic models of the capture process have been developed and validated by comparison with transient experimental data obtained from the pilot plant to subsequently study process and control system performance during load variations.

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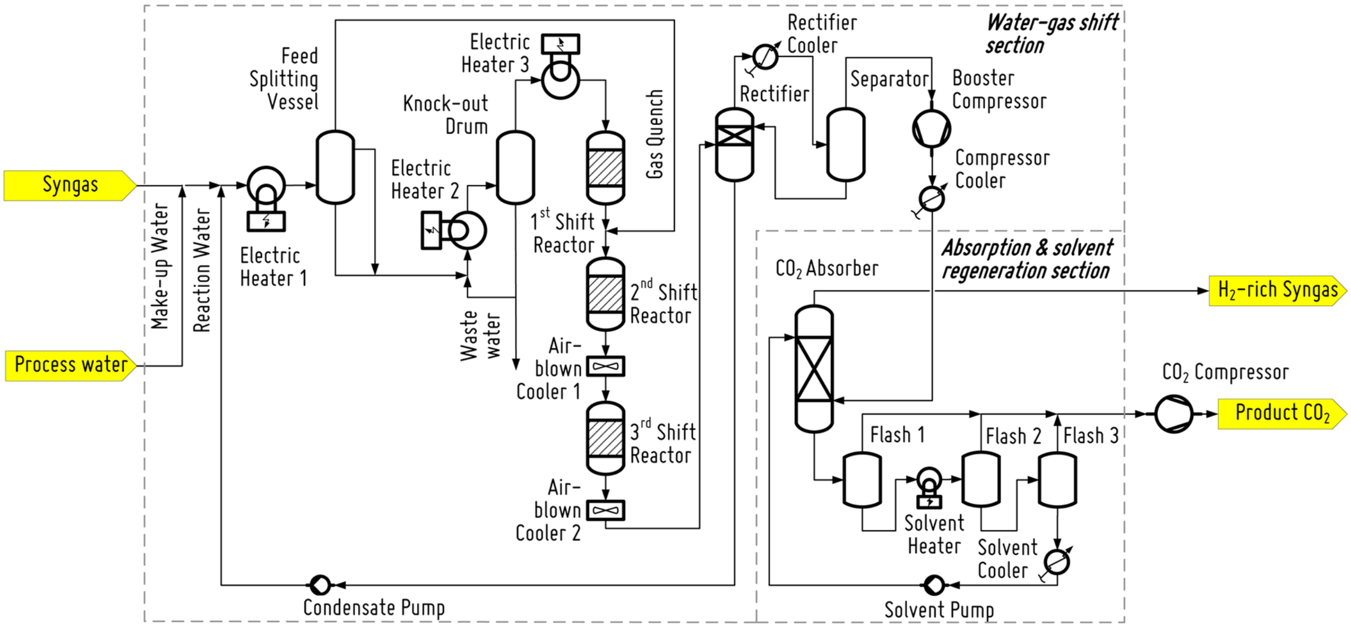

Figure 1. Process flow diagram of the CO_2 capture pilot plant.

The literature dealing with transient performance and control of IGCC power plants with integrated CO_2 capture units is scarce.[8−10] Few studies document model development and simulation of the IGCC processes without CO_2 capture,[6,11,12] and some authors focused only on dynamics of gasifiers[13−18] or auxiliaries, such as the air separation[19,20] or the fuel system.[21] Heil et al.[22] presented the modeling of a precombustion CO_2 removal unit utilizing methanol as physical solvent. The thermophysical properties of the fluid mixtures involved in the process were modeled with simplified thermodynamic laws and correlations.

Because of the unavailability of experimental data, both steady-state and dynamics, validation of CO_2 capture process models is still to be performed. The lack of validated process models is related to the fact that currently hardly any research project aimed at the demonstration of precombustion CO_2 capture technology is undertaken, resulting in a small number of pilot plants of this sort, namely Buggenum (Vattenfall),[7] Eagle (J-Power),[23] and Puertollano (Elcogas).[24]

In comparison, research in the field of postcombustion CO_2 capture is much more lively with a number of pilot and demonstration plants in operation.[25] However, data acquisition of these campaigns proved to be difficult due to challenges concerning the dynamic operation of postcombustion CO_2 absorption plants[26] and, as a consequence, only a few publications document also dynamic model validation.[27,28] Similar challenges affect also transient operation of the precombustion capture process, and therefore comprehensive experimental investigations accompanied by modeling activities to develop detailed and accurate dynamic models for process and control system design are still required.

The novel aspects of this work are as follows: (a) Demonstration of the application of a state-of-the-art, object-oriented modeling paradigm to the precombustion CO_2 capture process using a nonproprietary modeling language, which is tool-independent and which can be used with proprietary or open source simulation environments. The fluid properties are computed with accurate thermodynamic models, which have been implemented within an in-house property package (free for academic use). The model libraries containing the developed process models are made available for academic purposes under an open source license agreement. (b) Comprehensive dynamic model validation, whereby component, subsystem, and system models are validated by comparison with experimental data obtained from a pilot plant. Various open-loop and closed-loop transient tests were performed, by monitoring the response to stepwise changes of different operational parameters, such as syngas load, syngas composition and solvent mass flow rate. (c) The validated models have been used to investigate different control strategies aimed at improvement of the dynamic performance of the capture unit.

This paper is structured as follows: first the CO_2 capture process is briefly described in section 2. Model development is explained in section 3, covering the modeling approach, utilized tools, and a description of the Modelica models. The dynamic validation of the models including experiments and exemplary results are treated in section 4, while the process analysis focusing on control strategy improvement is presented in section 5. Finally, concluding remarks are given in section 6.

2. CO_2 CAPTURE PROCESS

The process flow diagram of the CO_2 capture pilot plant built at the site of the Buggenum IGCC power station is depicted in Figure 1. This plant is a simplified, smaller version of a foreseen large-scale capture plant, equipped with sensors and analyzers allowing for extensive performance measurements.[7]

The syngas from the gasifier, which contains about 55−60 mol % CO and 2−6 mol % CO_2, enters the water−gas shift (WGS) section of the CO_2 capture plant at process conditions of 21 bar and 40 °C and is mixed with process water (makeup) to obtain a preset steam/CO ratio (1.2 kg/kg). The syngas−water mixture is fully evaporated and superheated by means of three electric heaters. Carbon monoxide present in the syngas is converted into hydrogen and carbon dioxide via a three-stage, sweet, high-temperature water−gas shift process with interstage cooling (shift reactor inlet about 340 °C, shift reactor outlet about 480 °C for shift reactor 1 and 2, while about 370 °C for shift reactor 3). The partial bypass around the first shift reactor (gas quench from the feed split vessel) allows for lower steam consumption, hence substantial energy saving.[29,30] The excess process water is recovered from the shifted syngas through condensation in the rectifier and recycled.

After compression in the booster compressor in order to overcome the pilot plant pressure loss, the shifted syngas, which contains about 35−40 mol % of CO_2, enters the CO_2 absorption and solvent regeneration section. Carbon dioxide is removed from the syngas in a packed column (CO_2 absorber) by means of physical absorption utilizing the solvent dimethyl ether of polyethylene glycol (DEPEG) at process conditions of 40−45 °C and 21.5−22.5 bar. The resulting H_2-rich syngas is fed to the gas turbine of the combined cycle power plant, and the CO_2 is recovered by three-stage depressurization of the loaded solvent (flash pressures, which have been obtained by design optimization: 7.5, 2.9, and 1.3 bar). The lean solvent is recycled to the absorber, while the CO_2 product stream is compressed and, in the case of the pilot plant, mixed with the H_2-rich syngas. Typically, 80−85% of the CO_2 present in the shifted syngas is removed. A more detailed process description is given by Damen et al.[7]

The large-scale CO_2 capture plant process is very similar to the described process of the pilot plant, with the main difference being thermal energy recovery, or so-called heat integration within the WGS section: Electrical heaters and coolers are replaced by feed-effluent and feed-steam heat exchanges, whereby the steam is drawn from the heat recovery steam generator of the combined cycle power plant. In the absorption and solvent regeneration section, the gas recovered from the first flash vessel (also called H_2 recovery vessel), which primarily contains coabsorbed hydrogen, is recompressed and recycled to the absorber column. This way the combustible H_2 is not lost with the CO_2 product.

3. MODEL DEVELOPMENT

3.1. Modeling Approach. For the prediction of transient process performance, nonlinear dynamic models based on first-principles were developed following a modular approach in order to master system complexity, for example, the system is decomposed into suitable component models, which are connected through interfaces representing physical boundaries. Typically, zero-dimensional or one-dimensional component models were considered, which provide a sufficient degree of detail for accurate predictions of the transient system performance.

The models were implemented using the object-oriented, equation-based Modelica language.[31,32] Modelica is a non-proprietary modeling language, which is supported by various proprietary as well as open source simulation tools. Modeling features include, among others, reusability and extensibility, which allows an easy reuse of models developed by other researchers, or during previous projects, together with adaptation of existing models. Currently, an increasing number of open source and commercial Modelica libraries covering different engineering fields are available.

3.2. Thermophysical Properties. The thermophysical properties of the two-phase multicomponent syngas−water and sygnas−DEPEG mixtures are calculated with the perturbed chain-statistical associating fluid theory (PC-SAFT) equation of state (EoS)[33] due to its success in predicting vapor−liquid equilibria of complex fluids and mixtures for a broad range of conditions. For simplicity, the solvent DEPEG, which is a blend of glymes, is represented as a pseudopure fluid in the thermodynamic model. The fluid parameters are estimated following a method demonstrated by Nannan et al.[34]

This EoS has been implemented, together with fast and robust algorithms, into an in-house property package,[35] which is interfaced with the dynamic modeling tool. The use of external fluid property functions in Modelica process models imposes some restrictions to model development. Specific attention requires the formulation of the differential model equations, the choice of state variables, and the causality of the system model. A detailed discussion of these modeling aspects is beyond the scope of this paper, and the interested reader is referred to the publication of Trapp et al.[36]

3.3. Development of Component Models. The modeling of the CO_2 capture process requires various component models. Whenever possible available Modelica library models were reused. For example, basic component models such as sinks, sources, valves, pressure drops, pumps, heat exchange, and flow models, are taken from the ThermoPower library,[37,38] and adapted in terms of their media models, which have been replaced with functional calls to the external property tool.

New models were developed and implemented for the following components:[39,40]

Flash Vessel. The process of phase separation is modeled under the assumption of thermodynamic equilibrium between the liquid and vapor phase at all times. The model describes the holdup of vapor and liquid with conservation equations applied to control volumes containing the two-phase fluid in thermodynamic equilibrium. Saturated conditions are assumed for the liquid and vapor outlet streams, therefore entrainment of liquid in the vapor flow is neglected. The flash vessel model is implemented as a storage component, hence flow-friction losses are not considered. The static pressure head due to the liquid level in the vessel is accounted for in the algebraic momentum balance. Heat transfer from the fluid (both vapor and liquid phase) to the vessel wall, storage of thermal energy in the wall, as well as thermal energy losses to the environment are neglected. Superficial condensation is thus also assumed to be negligible.

Water−Gas Shift Reactor. The reaction of carbon monoxide with steam to produce carbon dioxide and hydrogen is described in a lumped-parameter model. The syngas entering and leaving the reactor is an ideal gas mixture containing CO, CO_2, H_2, H_2O, and N_2. Other trace constituents are neglected. The model accounts only for the WGS reaction. Intermediate reactions involving other chemical species are neglected. The reactor model is subdivided into five submodels: reaction node, mixing gas volume, convective heat transfer, thermal storage, and pressure drop. The object diagram of the model is depicted in Figure 2.

The WGS reaction takes place in an infinitesimally small volume (reaction node) representing one finite discretization of the catalyst, and it is assumed that it reaches thermodynamic equilibrium. The storage of mass and energy in the bulk phase of the reactor are modeled in a perfectly mixed volume (mixing gas volume) which receives the reaction products. This control volume exchanges heat with the catalyst by means of convection. The storage model describes the storage of thermal energy in the catalyst. Heat transfer to the environment is neglected.

The water−gas shift reactor is discretized in the axial direction by an array of reactor models in order to correctly

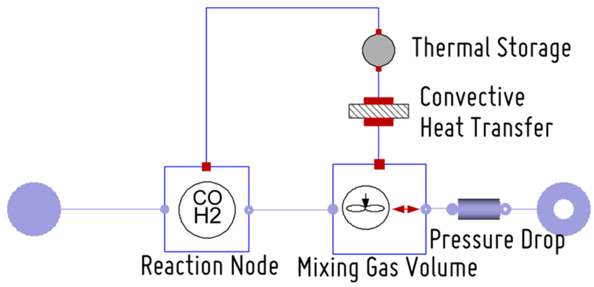

Figure 2. Object diagram of reactor component.

describe the gradual changes in reactor outlet conditions during transient operation. Changes in the reactor inlet conditions reach the reactor outlet with a delay due to thermal storage in the catalyst, which cannot be represented with a 0-dimensional model due to the high number of transfer units between the gas and the catalyst itself.

This one-dimensional discretization does not however represent the actual axial reactor profile as equilibrium conditions are assumed in each reactor model element for simplicity. At steady-state conditions the equilibrium temperature is reached at each discretization of the catalyst, which also determines the temperature-dependent WGS reaction.

Pilot Plant Specific Heater and Cooler Components. Various electrical components for evaporation, superheating, cooling, and condensation were in particular developed for the pilot plant, and will not be part of a large-scale plant with this specific configuration. The models were typically subdivided, if applicable, in flow models, heat transfer models, and thermal storage models. Whenever possible models from the ThermoPower library were used, typically in case the medium was water or ideal gas, or adapted. The heat transfer coefficients were either computed with specific heat transfer correlations or tuned to experimental data.

Absorption Column and Sump. The model of the packed column for physical absorption (no chemical reactions) is discretized in theoretical stages in the axial direction, and counter-current flow of the vapor and liquid is assumed. Each stage is modeled by an equivalent tray module (only storage model) together with a resistive module. These stages are connected in series to form a column model as shown in Figure 3.

In the equivalent tray module, pressure, temperature, and composition of the liquid and vapor phase are determined by solving the conservation equations for mass and energy assuming thermodynamic equilibrium between liquid and vapor.[41] This module is based on the flash vessel model with the same assumptions. In the resistive module, the momentum equation is substituted by empirical correlations to describe the hydrodynamics of the stage predicting the liquid and vapor flow rate as a function of the pressure difference between the stages, the liquid holdup, and the packing characteristics. Empirical correlations for the pressure drop and liquid holdup are, for example, those of Stichlmair et al.[42] and of Billet and Schultes.[43]

The sump model is implemented as a storage component accounting for the holdup of liquid. The static pressure head due the liquid level in the sump is modeled with an algebraic momentum balance. Storage of thermal energy in the sump wall and heat losses to the environment are neglected.

The subsystem models (e.g., water−gas shift section, absorption and solvent regeneration section) and system model of the capture plant are assembled by connection of individual component models. Component as well as system models are implemented in a Modelica library which is made available for academic use.

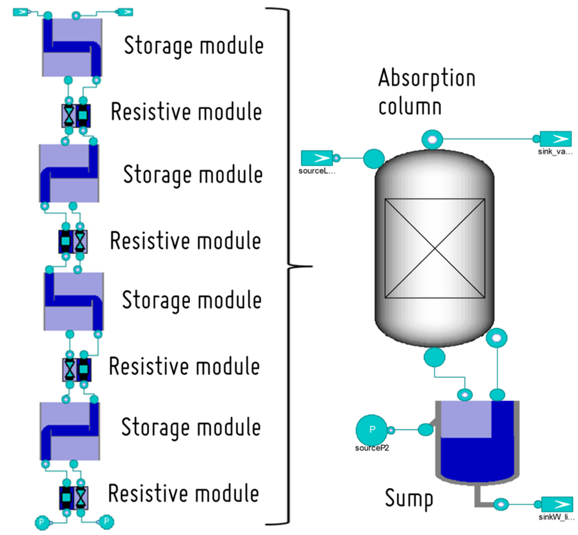

Figure 3. Model structure of the absorption column and sump.

4. DYNAMIC MODEL VALIDATION

In general terms, the dynamic model validation discussed here aims to demonstrate whether the developed CO_2 capture process models, with their identified relevant phenomena and, assumptions and hypothesis, are a sufficient representation of the actual process in relation to the purpose of the model. The accuracy of the simulated process transients must therefore be sufficient in order to perform control-strategy design and to improve the dynamic operation of the plant.

4.1. Validation Approach, Experiments, and Results. The fully instrumented pilot plant was designed for operational flexibility to investigate the influence of various operating conditions, both steady-state and dynamic, on component and system performance, and therefore facilitated comprehensive model validation. Moreover, with the support and knowledge of the experienced plant operators, a wide range of different experimental tests could be executed. These possibilities allowed dynamic validation at three different levels: component, subsystem, and system level.

In a first step, the individual, newly developed component models of the CO_2 capture process were validated, such as the water−gas shift reactor, various heater and cooler components, and the absorber column. For each component, open-loop and closed-loop experiments were performed. During open-loop tests process dynamics are analyzed without the control system in operation, whereas in closed-loop tests, process and control system performance are assessed for a realistic operational scenario.

The open-loop tests (e.g., step responses) are suitable to reveal the inherent dynamics of the process and make sure that it is correctly captured by the dynamic model. Unfortunately, they cannot always be performed safely, or conveniently, on the real plant. Closed-loop tests are easier to carry out, but only provide relevant dynamic information in the frequency range around the controller's crossover frequency. On the one hand, this might in fact be better than open-loop tests: if the controller is very fast, the closed-loop behavior depends on the fast dynamics of the process, which might be hard to discern

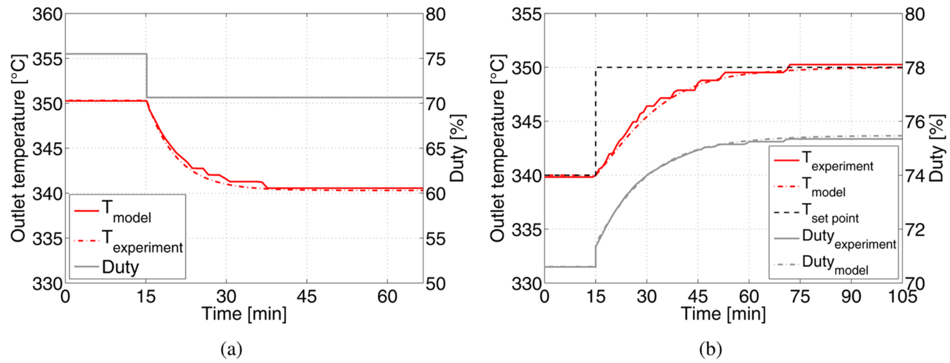

Figure 4. Validation of superheater component (see Figure 1). Comparison of measurements and simulation results for outlet temperature and duty: (a) open-loop test, step decrease in heater duty; (b) closed-loop test, step increase in temperature set point.

clearly from open-loop tests, such as step responses. On the other hand, if the controller installed on the plant is very conservative (i.e., slow), then closed-loop tests might fail to reveal the dynamics of interest in case more aggressive controllers are designed, based on the model. In general, a combination of both tests is the best option in order to comprehensively assess the quality of the model.

The linear system response was evaluated by applying small perturbations to the input variables starting at steady-state operation, though large enough to obtain an acceptable signal-to-noise ratio. For relatively small changes the expected response of an upward and downward step change is symmetrical in terms of the main dynamic parameters of the transient, such as time and value of maximum overshoot, settling time, presence, and damping of oscillations, etc.

During transient operation all input variables other than the perturbation variable should be maintained constant. In case this is not possible because of process limitations or other uncontrollable disturbances, such as, changes in environmental conditions, then these variables are prescribed inputs for the dynamic model.

The individual CO_2 capture process components were validated following the outlined procedure. The validation of a superheater component model is briefly described here as an example.

Example for Component Validation: Superheater. The main phenomena modeled by the electric superheater component, see Figure 1, are the storage of thermal energy and the heat transfer. The best manner to demonstrate that these phenomena are modeled with sufficient accuracy is by analyzing if the superheater performance, in terms of response in outlet temperature due to a change in heat duty, is satisfactorily reproduced by the model. The open-loop and closed-loop experiments were designed such that the inlet conditions in terms of mass flow, temperature, pressure, and composition remained constant during the experiment. This required the operation of some upstream control loops in manual mode.

During the open-loop experiment a downward step was applied directly to the heater duty without the PI controller in operation. The step in power corresponds to a change in the superheater outlet temperature of approximately 10 °C. During the closed-loop experiment the superheater temperature set point was changed stepwise by 10 °C. The temperature controller reacts by adjusting the duty in order to reach the desired set point value. The model validation is performed by comparing measurements of the superheater outlet temperature transient as response to the applied perturbations to simulation results obtained with the superheater component models.

Figure 4a shows the results of the temperature transient for the open-loop step change in superheater duty. A satisfactory match between the measurements and the model predictions is observed considering the main dynamic parameters. The temperature measurements are not smooth and the steplike changes occur because the measurement is recorded if the variable change in absolute value exceeds a defined and adjustable threshold. This threshold-based mechanism might on the other hand mask the appearance of noise in the measurements. In Figure 4b the closed-loop transients of the temperature and of the heat duty are compared for a step increase in the temperature set point. For both, the process and the control variable, simulation results and experimental data show good agreement. The measurements of the heat duty are much smoother than the temperature recordings.

In a next step, subsystem models were developed on the basis of the validated component models. A subsystem, such as the water−gas shift section or the absorption and solvent regeneration section, cannot be operated without control because of plant safety and stability. Therefore, partial open-loop experiments were designed, whereby only control loops which do not compromise the safe operation were put into manual mode. In the following, the validation of the water−gas shift section model is discussed as an example for subsystem validation. The detailed model validation of the absorption and solvent regeneration section model is demonstrated in a related paper.[44]

Example for Subsystem Validation: Water−Gas Shift Section. During the partial open-loop experiment an upward step change was applied directly to the syngas control valve without the syngas controller in operation. All other controllers of the WGS section were kept in automatic mode and are summarized for a better understanding in Table 1. The step in valve opening corresponds to a change in syngas flow of approximately 100 kg/h in absolute and 10% in relative terms. The scheme of the subsystem model used for the dynamic simulations is depicted in Figure 5.

The pressures at the syngas inlet, at the reaction water valve inlet, and at the reactor 3 outlet are input variables of the dynamic model. The first two pressures can be set constant for the simulation. The back pressure of reactor 3 varies during the experiment; therefore, the actual pressure measurements, which are depicted in Figure 6a, are provided as model input.

The perturbation was applied at t = 50 min starting from an initial syngas load of 1000 kg/h. Owing to the step-opening of

Table 1. Control Loops within the WGS Section (see Figure 5)

controlled variable	control variable	set point
syngas mass flow rate	opening of syngas control valve	1100 kg/h[a]
outlet temperature of heater	heater duty	172 °C
level of 1st vessel	opening of water control valve	1300 mm
level of 2nd vessel	opening of liquid control valve of 1st vessel	800 mm
mass flow rate of reactor 1	reboiler duty	internally calculated[b]
inlet temperature of reactor1	superheater duty	340 °C
inlet temperature of reactor 2	set point for opening of quench control valve	340 °C
inlet temperature of reactor 3	fan speed of cooler	340 °C

[a]For the presented test run this control loop was operated in manual mode, hence the PI controller is not depicted in Figure 5. [b]This loop is used to control the ratio of water/syngas (set point) by calculating the expected mass flow rate of reactor 1 based on the measured syngas and quench flow.

the syngas control valve, the mass flow rate of reactor 1 increases almost instantaneously pulling more syngas−water mixture from the downstream components, see Figure 6b. This results in a fast increase in the syngas inlet flow, Figure 6d. Furthermore, the increased mass flow rate of reactor 1, now containing a larger total amount of thermal energy, causes an initial increase of the inlet temperature of reactor 2, see Figure 7c, when mixing at the outlet of reactor 1 with the quench flow, which remained rather unchanged in terms of flow and temperature during this initial transient. The quench flow control opens the quench valve (Figure 5) in order to maintain the inlet temperature of reactor 2. This causes an increase in the quench flow and decrease in the mass flow rate of reactor 1. The operating condition of the system starts to fluctuate, whereby almost all process variables are influenced. The controllers stabilize the operation such that the oscillations are dampened and the new steady-state is approached at t = 200 min.

The model predictions for the mass flow rate of reactor 1, of the quench stream and syngas stream compare well with the experimental results (see Figure 6), in particular the initial response in terms of rise time and maximum overshoot are predicted accurately. The presence of oscillations is captured by the model; however, the damping is overestimated, and this is discussed in more detail in the following. Also for the temperature of the quench and the inlet temperature of the superheater a satisfactory agreement between the simulation results and the measurements is achieved in terms of the main dynamic parameters (see Figure 6e,f).

Figure 7 shows the comparison of model predictions and measurements for the outlet temperature of reactor 1 and reactor 2. Oscillations are observed in the experimental results especially for the outlet temperature of reactor 2, and these are mainly caused by oscillations in the inlet temperature of reactor 2, see Figure 7c. Fluctuations in the composition of reactor 2 have negligible impact. The inlet temperature of reactor 2 is influenced by changes in flow rate and/or temperature of the outlet stream of reactor 1 and the quench stream and is controlled by a master-slave temperature controller damping the oscillations. These oscillations are not inherent to the process dynamics alone, but are rather the result of the interaction between the controller dynamics and the process dynamics, which is therefore captured correctly in the frequency range which is relevant to closed-loop performance. The damping of the oscillations is a bit overestimated by the process model, which means that additional relatively small unmodelled delays (such as, for example, those due to sensor and actuator dynamics) are present on the real plant.

Regarding the outlet temperature of reactor 1, the amplitude of the variations is not predicted accurately by the model. The maximum overshoot is underpredicted by 8 K. However, the general dynamic trend, the presence of oscillations, and the settling time can be deemed in good agreement with the experimental data. The results of the comparison are similar for the outlet temperature of reactor 2 with maximum deviations of 10 K. The mismatch in the reactor outlet conditions originates from the use of a simplified reactor model which assumes equilibrium conditions throughout the reactor. To correctly

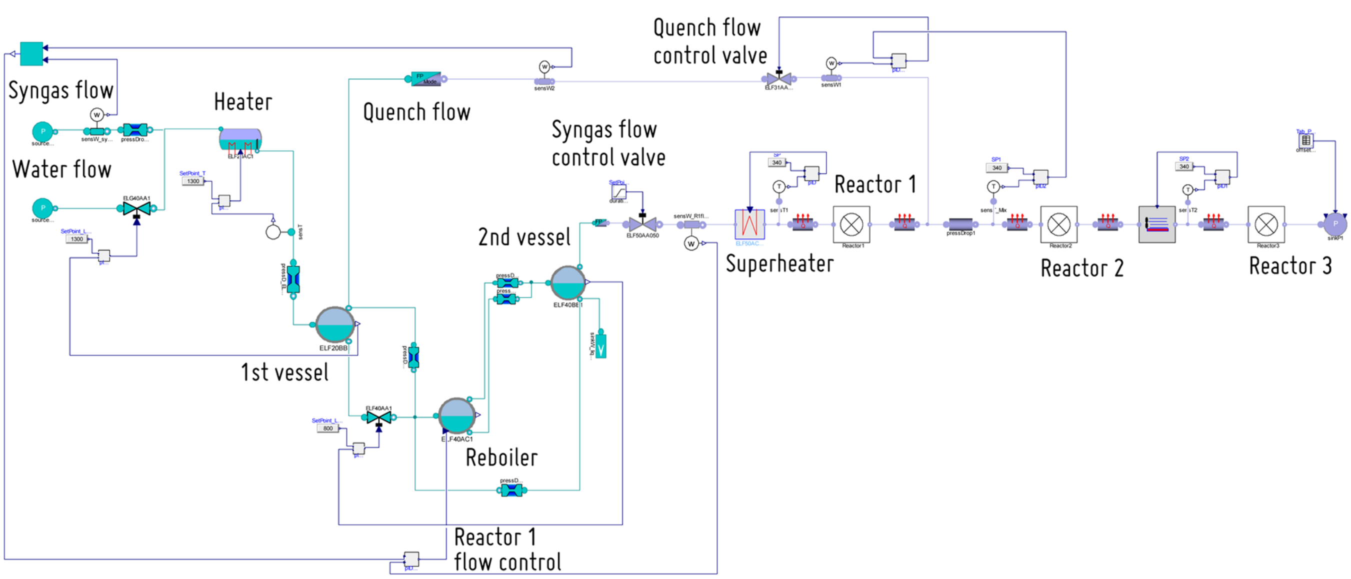

Figure 5. Object diagram of the water−gas shift section model (rectifier, separator, and booster compressor not included).

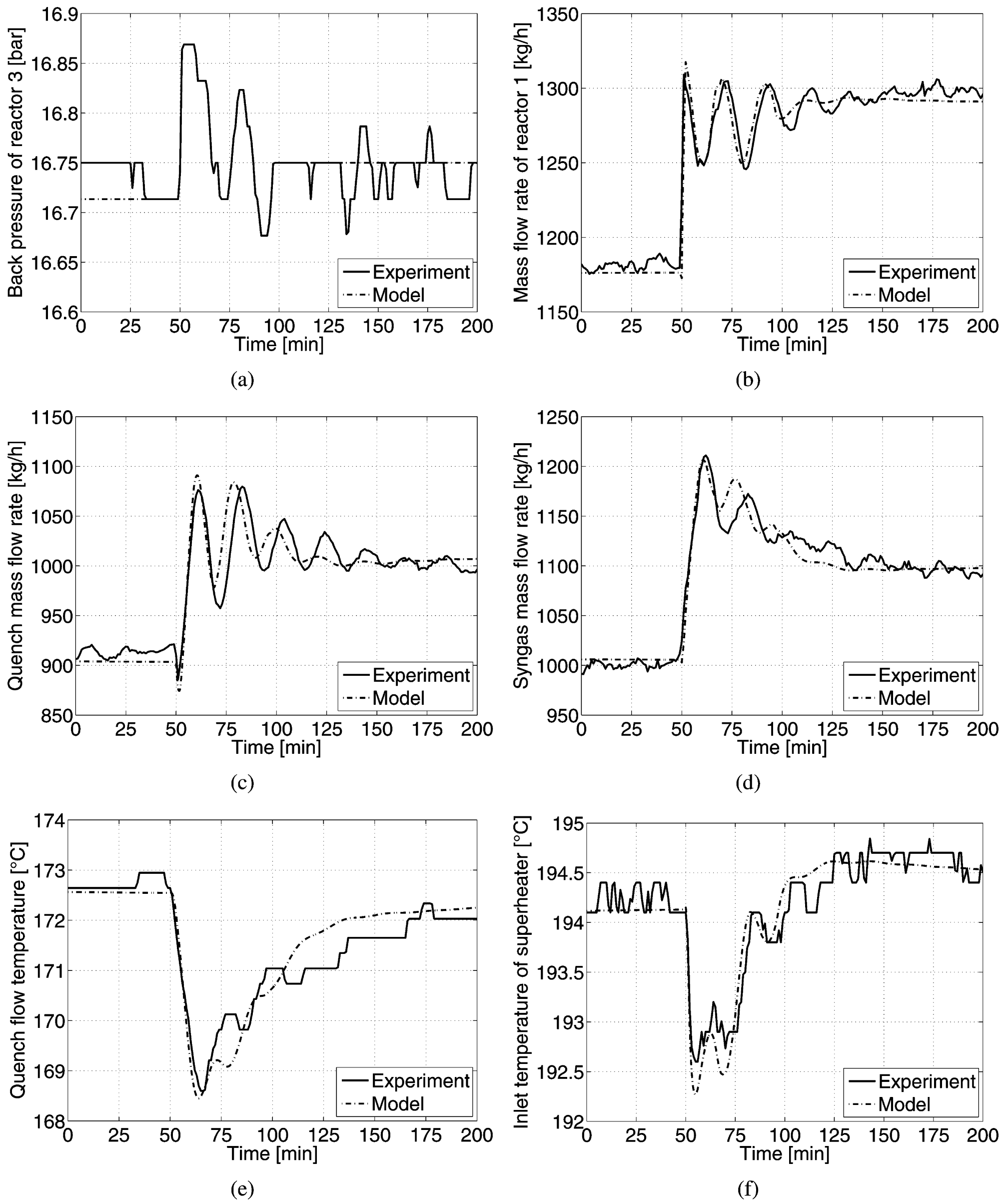

Figure 6. WGS section model validation: comparison of measurements and simulation results for a step increase in syngas control valve opening (part I).

predict the reactor performance, a more computationally expensive, kinetic-based model would be required. This model would allow the accurate prediction of the axial reactor temperature profile. It shall be kept in mind, that in realistic load change scenarios the perturbations are gradual and not stepwise as in the partial open-loop experiment. Therefore, the system response will be smoother and the amplitude of the variations smaller. As far as the reactors are concerned, this would lead to a better agreement between the actual transients and the simulations results.

For the purpose of analyzing the overall transient performance of the capture plant and evaluating different control strategies, the use of a simplified reactor component model as part of the entire dynamic model is arguably sufficient. In case the dynamic model should be used to fine-tune the control parameters of the plant control system, a more detailed reactor model might be required.

To summarize, the initial and final steady-state values of the main process variables are reproduced with less than 1% deviation with respect to measured values, an accuracy that can be considered satisfactory. Good steady-state predictions are also achieved for validation at off-design operation (60% syngas load). This similar validation case is not included here for the sake of conciseness. Considering the main dynamic characteristics of the observed transients, namely, time and value of maximum overshoot, settling time, frequency and damping of oscillations, they are predicted within 20% of the value that can be obtained from experimental data; larger errors are found in the temperatures of the reactors, and they can be attributed to the less accurate predictions for fast transients by the simplified, equilibrium-based reactor model.

Finally, the validated subsystem models were combined to form a system model of the CO_2 capture process. For system model validation a closed-loop assessment of the process and control system performance was carried out considering

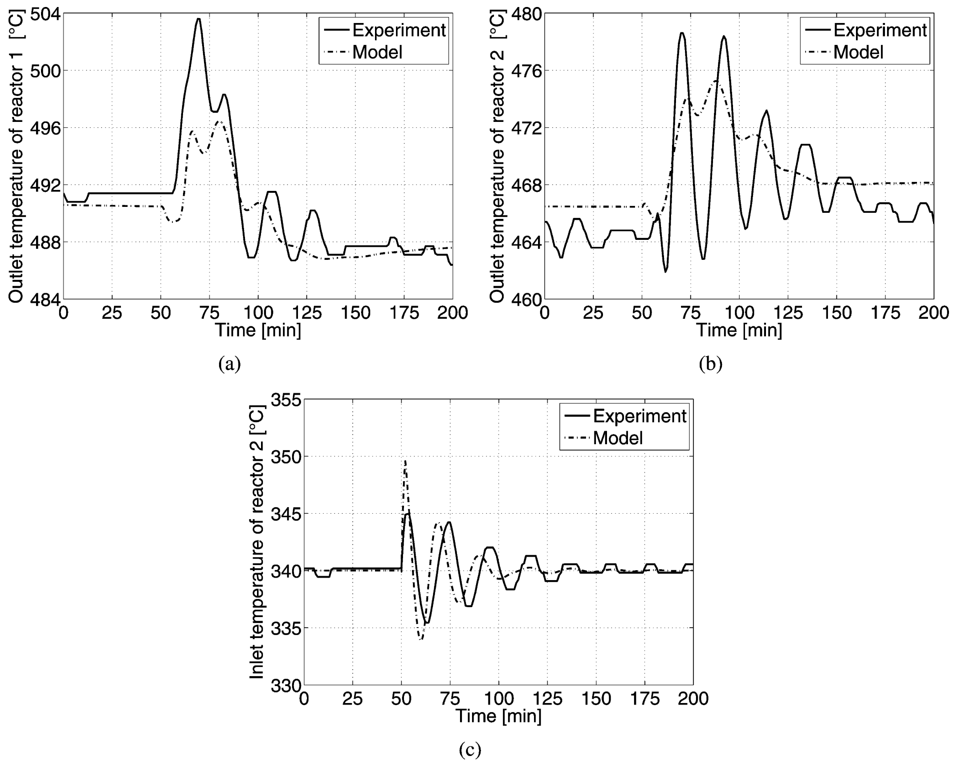

Figure 7. WGS section model validation: comparison of measurements and simulation results for a step increase in syngas control valve opening (part II).

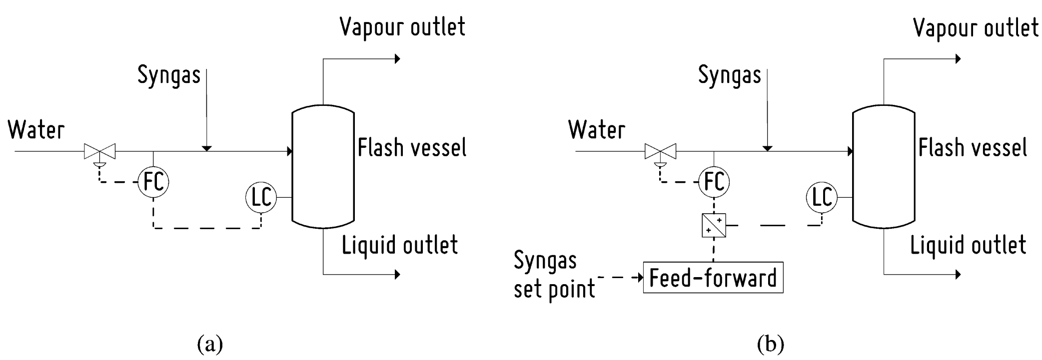

Figure 8. Flash vessel level control via (a) cascade and feed-back control; (b) cascade, feed-back, and feed-forward control.

changes in syngas load. To avoid repetition, the results are not discussed in detail but presented together with the control system analysis in the next section.

Summing up, this step-by-step validation approach from component to subsystem to system level provides (a) validated and reliable component models, which can readily be integrated into other processes (adaptation of geometrical data required), (b) validated system models including process control, and (c) detailed understanding of the capture process in relation to which phenomena in the individual components are relevant and require accurate modeling to serve the purpose of dynamic process analysis and control system design.

5. PROCESS ANALYSIS

The dynamic performance of fossil-fuelled power plants becomes increasingly important, and hence an integrated capture process must be able to follow relatively fast load variations; however, it might also be required to temporarily only reduce the load of the energy-intensive CO_2 capture process, while maintaining the gasifier load for an IGCC power plant. For example, this can be the case when the market demands more energy or it is economically more favorable to produce energy instead of capturing CO_2. The amount of CO_2 capturing and hence its energy consumption might also be adjusted for primary or secondary frequency control. The control system of the CO_2 capture unit should therefore allow frequent and prompt load variations.

The objective of this analysis is to study the improvement of the dynamic performance of the precombustion CO_2 capture unit by investigating an improved control strategy and demonstrating, by means of dynamic simulation, that this strategy might work if implemented into the actual plant control system. The decentralized control system based on PI controllers as implemented in the pilot plant is used as reference (in the following referred to as *reference control*) to evaluate the improved control. It needs to be mentioned that the pilot plant control is possibly far from being optimal and

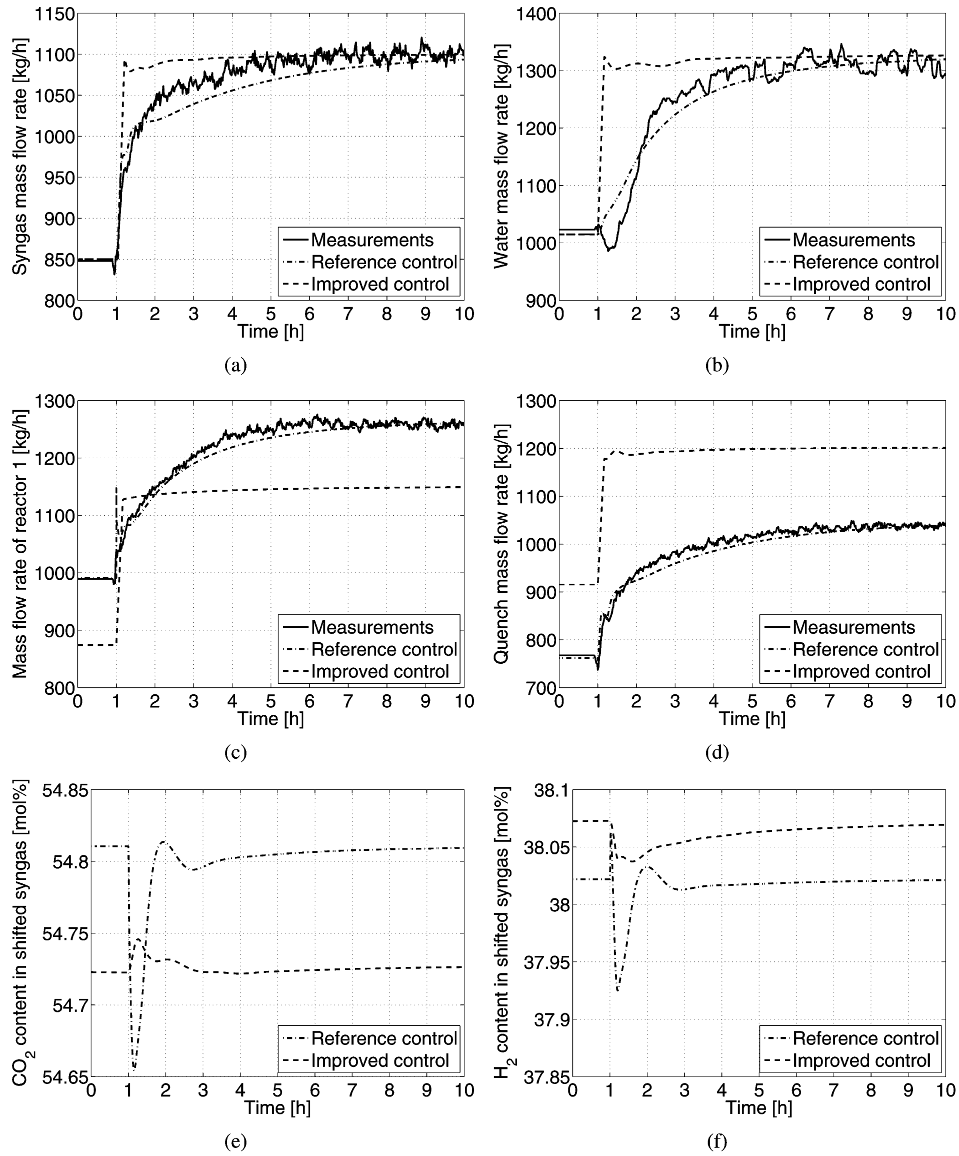

Figure 9. Comparison of simulation results: reference versus improved control. The objective is to change the operating condition from partial-load to full-load (part I).

was designed and tuned in order to achieve stable and safe operation during the test campaign.

For the investigation of control strategies the validated dynamic system model of the CO_2 capture pilot plant was used. The design of the pilot plant and of a foreseen large-scale plant are very similar, with the main difference concerning heat integration in the WGS section. Therefore, it can be assumed that a developed control strategy tested with the pilot plant model can also be applied to a large-scale plant with appropriate modifications.

Partial-load and full-load operation of the CO_2 capture unit differs mainly in terms of flow rates, whereas most of the other process variables and parameters are kept to the same values. To allow for fast load variations it is particularly important to apply good set point management for the temperatures in the WGS section, as the thermal inertia of the system is much larger than the mass inertia. Considering the current control architecture, the only ways to drastically improve the control performance is to introduce a centralized controller and/or feed-forward action. Given the nature of the process, feed-forward control leads to a simpler control architecture, which is easier to design and also easier to understand for the plant operators. Hence, a control strategy based on feed-forward, feed-back, and cascade control was implemented into the pilot plant model, and tested for virtual plant operation. The improved control strategy and its implementation is explained in more detail in the following.

Figure 8a shows an example of cascade control, as implemented in the dynamic model. The cascade control consists of two control loops. The master control compares the level measurement (process variable) with a given level set point and changes the set point of the slave control (control variable of the master control). The slave loop compares the flow measurement (process variable) with the set point provided by the master loop and changes the valve opening (control variable) accordingly. The advantage of cascade control is that it allows the system to be more responsive to disturbances, and it is particularly useful for systems with long dead and lag times.[45] However, this comes at the cost of higher system complexity and requires more process instrumentation.

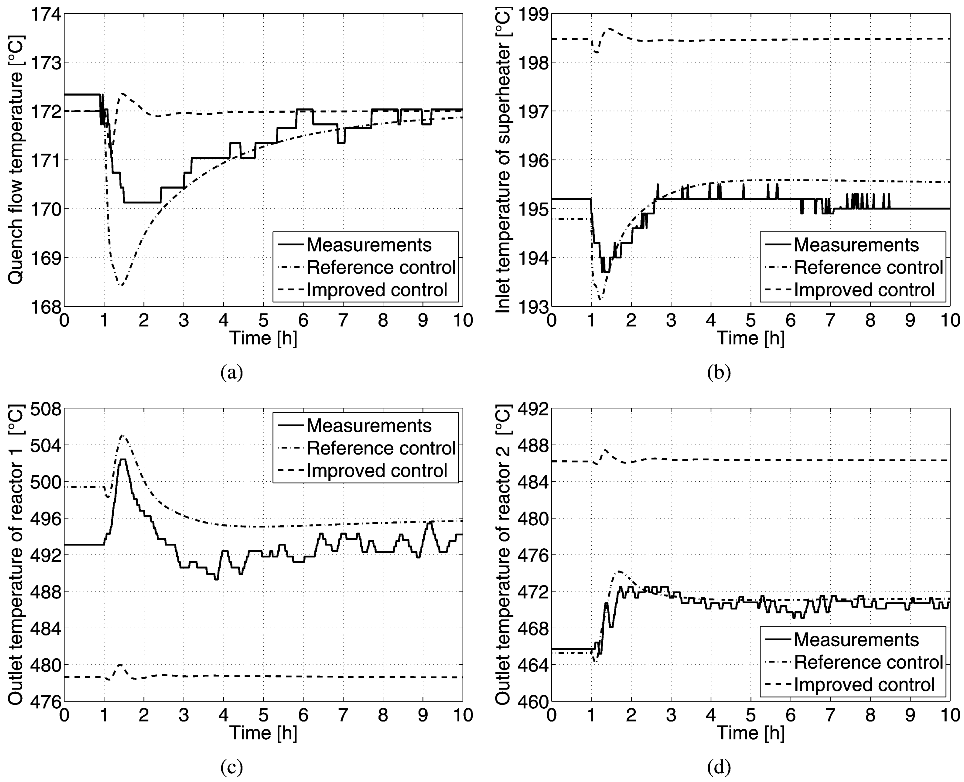

Figure 10. Comparison of simulation results: reference versus improved control. The objective is to change the operating condition from partial-load to full-load (part II).

A cascading control scheme has been implemented in the WGS section for the control of the liquid level in both vessels, the syngas mass flow rate and the inlet temperature of reactor 2.

Figure 8b shows the application of feed-forward control to the reaction water loop as a paradigmatic case. The aim of feed-forward control is to measure disturbances upstream of the system and compensate for them before the system variables deviate from the set point. In the case of the capture unit, one of the main disturbances is the change in syngas load. Hence, the feed-forward control receives the syngas set point as input and determines the set point for the water flow control based on an explicit equation. The feed-back control ensures that the level set point is maintained. The advantage of feed-forward control in comparison to feed-back control is that disturbances do not need to propagate through the process in order to take control actions, hence the set point control is more accurate. However, to accurately predict feed-forward control actions, accurate measurements and adequate disturbance predictions are required. In the case of nonideal processes, accurate prediction might require models consisting of nonlinear equations. The feed-forward control logic has been implemented in the dynamic model for all master control loops. The feed-forward action is determined on the basis of the syngas set point (it is currently based on a simple linear correlation), and is sent together with the feed-back action to the slave control loop. The coefficients of the linear equations are determined by considering two steady-state operating points. The implementation can easily be extended: for example, in case real plant data are available, more elaborate correlations can be tuned to cover a wide operating range.

Observation of the results of experimental tests on the pilot plant show that the steady-state reactor performance varies from partial-load to full-load, which is revealed by the different values of the reactor outlet temperatures. This hampers fast load (i.e., syngas flow) changes of the plant by means of feed-forward control, because long settling times of the large thermal inertias of vessels and reactors slow down the system response. Feed-forward control could be more effective if such performance changes were eliminated, or at least greatly reduced.

The cause of the performance difference is related to changes in the thermodynamic state of the second vessel upstream of the reactors, in terms of temperature and pressure, which subsequently leads to changes in the inlet composition of reactor 1 and 2. This applies to the case in which the syngas inlet composition is constant, which is a justified assumption if the same type of fuel is used for gasification and if the gasifier remains at constant load.

In the case of the pilot plant, the inlet pressure of the capture unit is constant, and hence the actual vessel pressure is a result of the difference between the inlet pressure and the flow-dependent frictional losses. To maintain the vessel pressure at different loads, a pressure controller needs to be added.

Furthermore, the vessel conditions at part-load and full-load differ because of changes in process heat losses. In the pilot plant large heat losses occur at the inlet and outlet of the reactors because the reactor casing is overdimensioned for the installed amount of catalyst; furthermore, there are significant heat losses in the piping. These losses can be reduced by adding more insulation, which can be modeled by a reduction of the heat loss coefficients in the system model. This hardware modification will probably not be required in the full-scale plant, because of more accurate design and smaller heat transfer area to volume ratio.

Summing up, a much faster control performance can be achieved by implementing cascade controllers with slave flow controllers, by adding suitable static feed-forward actions to the

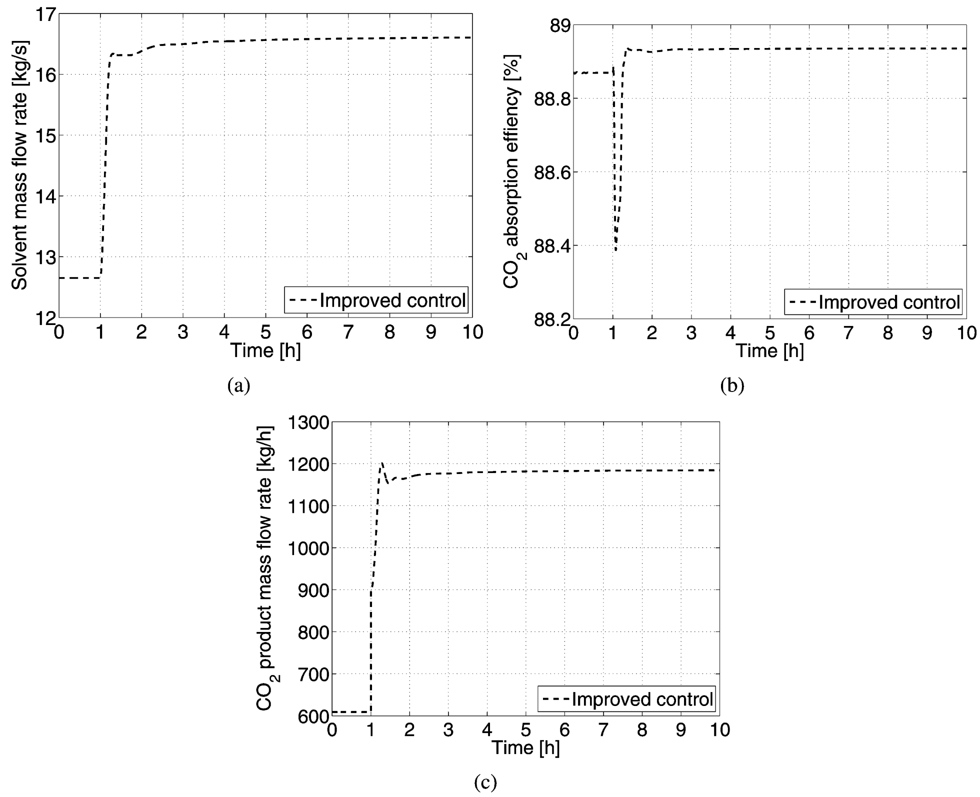

Figure 11. Simulation results of improved control. The objective is to change the operating condition from partial-load to full-load (part III).

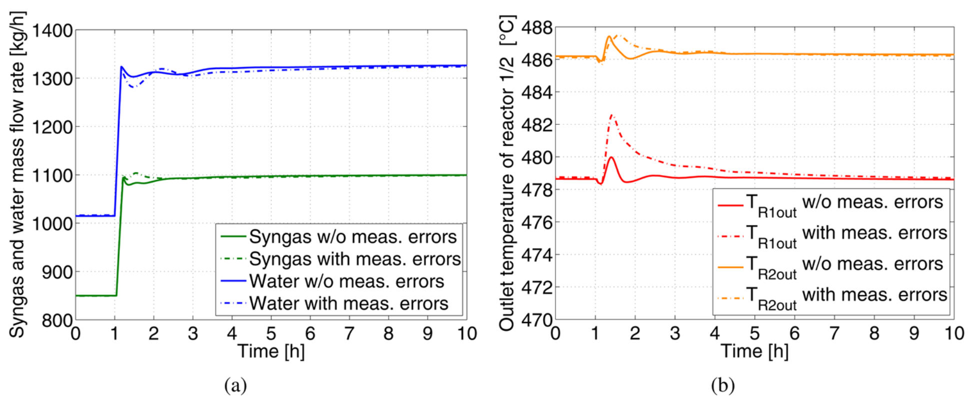

Figure 12. Comparison of simulation results: improved control with and without measurement errors. The objective is to change the operating condition from partial-load to full-load: (a) Syngas and water mass flow rate; (b) outlet temperature of reactor 1 and 2.

master controllers, by improving the insulation of lossy components, and by adding a pressure controller, the last two measures being necessary to avoid temperature swings when changing the load.

Figure 9 (process mass flow rates and shifted syngas composition) and Figure 10 (process temperatures) visualize the comparison of simulation results for the WGS section of the CO_2 capture plant obtained with the reference and the improved pilot plant control for a load variation from partial-load to full-load. The model predictions of the reference control have been validated with experimental data, as seen in the plots. In the simulation with the improved control, the syngas mass flow rate is ramped from 850 to 1100 kg/h in 10 min, based on expected future load change scenarios. In the simulations with the reference control, the syngas mass flow set point is changed instantaneously and the control system takes care of the load change. The dynamics of the reference control are rather slow; hence, there is hardly any difference between applying a 10 min ramp or a step, as will be demonstrated in the following. Therefore, the slightly faster step change was applied.

From the comparison of the reference and the improved control it can be observed that it is possible to subject the capture process to prompt load changes. The simulation of the process regulated by the improved control system indicates that, as far as the mass flow rates in the WGS are concerned, more than 95% of the final steady-state value can be reached within the ramping time. The vessel and reactor temperatures settle within approximately 60 min after the beginning of the perturbation. In addition, with the improved control (cascade and feed-forward) and the measures to maintain the reactor performance, the maximum overshoot during transient can be reduced. In the case of the improved process control, due to the change in heat losses and vessel pressure, the steady-state values of most of the variables are different in comparison to the simulation results obtained with the reference control.

Figure 11 visualizes the simulation results related to the absorption section in case the improved control strategy is adopted. The operators of the pilot plant manually adjusted the solvent mass flow rate in order to account for syngas load variations such that the performance in terms of CO_2 capture was maintained. No meaningful comparison to the reference control can therefore be provided. The implementation of the feed-forward control suggests that this strategy is a good replacement of the manual operation.

A ratio controller is implemented into the dynamic model of the plant in order to maintain the weight-based liquid-to-gas ratio of the absorption column, which allows the CO_2 removal efficiency to remain approximately constant for the operational range of the absorber. This has also been verified during tests at the pilot plant. Consequently, if the mass flow rate of the shifted syngas changes, the solvent mass flow rate is adapted accordingly. The results indicate that the absorption section quickly responds to fast load variations.

As mentioned, feed-forward control requires process measurements in order to predict the control actions. It is therefore important to investigate the impact of possible measurement errors on the performance of the proposed control strategy. Multiple simulations have been performed whereby a relative error of either +5% or −5% was applied randomly to each measurement. Figure 12 shows the comparison between a simulation unaffected by errors and one in which the errors have been applied in order to produce the largest variation in the main process variables. For conciseness, only four process variables are reported as representative examples for the difference in overall performance. When including measurement errors during the simulation, the overshoot and settling time increases, but only slightly for most process variables. The worst response is observed in the outlet temperature of reactor 1, where the overshoot more than doubles because of the measurement errors. This demonstrates that the performance of the proposed control strategy can decrease if measurement errors are present; however, the gain in improvement of the dynamic performance in comparison to the reference control is still significant.

By implementing the improved control system into the pilot plant it would be possible to verify if the predicted performance can be achieved and if there are phenomena which are not yet included in the model but have an impact on the control performance. Unfortunately, the test facility was shut down shortly after the test campaign because of economic reasons and a strategy change of the funding company, therefore the improved control could not be tested.

To conclude, it has been demonstrated that validated physical-based dynamic process models ease the design and testing of control strategies and that prompt load variations can be performed with a precombustion CO_2 capture unit. The qualitative results from this investigation related to the pilot plant model can be applied for the design of control strategies of a large-scale capture plant. Ultimately, the goal is to study the transient interaction between the CO_2 capture unit and the main power plant during load variations, which is beyond the scope of the work presented here.

6. CONCLUSIONS

This paper discusses the dynamic modeling and simulation of precombustion CO_2 capture plants for the use in control design. The models follow an object-oriented modeling paradigm. The models of the main process components, such as the flash vessel, the water−gas shift reactor, and the absorption column are presented briefly. Comprehensive model validation has been performed at component, subsystem, and system level by comparing dynamic simulation results to experimental measurements obtained from various open- and closed-loop transient tests performed at the CO_2 capture pilot plant operated at the Buggenum IGCC power station. Dynamic simulations with the validated system model have been used to investigate a promising control strategy based on feed-forward, feed-back, and cascade control to improve dynamic performance. The following conclusions can be drawn from this study:

- Transient performance in terms of prompt syngas load variations is a feasible operating mode for precombustion CO_2 capture plants featuring sophisticated control systems. This enables the IGCC power plant to respond to fast changes in the energy demand by reducing the load of the energy-intensive CO_2 capture process instead of adjusting the gasifier load.
- Physical-based dynamic models can be of manifold use during the design phase of new plants to evaluate different configurations, to support equipment selection and sizing, and to develop and test control strategies with the underlying aim to improve dynamic performance. A demonstration of these capabilities has been given in this paper with respect to control optimization. Moreover, the design and tuning of controllers prior to plant construction can save time and trouble during commissioning.
- The validated component and pilot plant system models have been implemented in an open source library, which can serve as a reliable foundation for the development of large-scale system models of a precombustion capture process. This requires the assembly of the required process models, by reuse of existing models and adaptation of equipment sizing along with the implementation and tuning of a control system. The flexibility of the object-oriented modeling approach allows an easy extension of existing models and implementation of more sophisticated model versions if required. Moreover, the model of the CO_2 capture plant can be easily integrated into models of gasification units and combined cycle plants in order to form system models of the entire IGCC power plant,[6] which can ultimately be used to study the interaction of the different units during transient operation.
- Finally, it is worth pointing out that the nonproprietary Modelica language is supported by various software tools, which has the advantage of not being bound to a specific proprietary simulation environment and gives the flexibility to explore the simulation capabilities of different tools. Moreover, it has been demonstrated that Modelica process models can be interfaced with external fluid media libraries for the computation of thermophysical properties. This allows an easy change of the process fluids as typically a wide range of pure fluids and fluid mixtures described with suitable and accurate equation of states are available in property packages. In

addition, the same property package can be interfaced with a wide variety of engineering software tools (e.g., steady-state and dynamic system modeling, component design, CFD, etc.) allowing for the use of the same thermophysical properties, thus eliminating one common source of uncertainty.

Future work might tackle, apart from the analysis of the transient performance of decarbonized IGCC power plants, also the dynamic optimization of load change trajectories, whereby the main challenge will be related to the system complexity.

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Notes

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REFERENCES

(1) *CO_2 Emission from Fuel Combustion*; International Energy Agency: Paris, France, 2012.

(2) *World Energy Outlook 2012*; International Energy Agency: Paris, France, 2012.

(3) Damen, K.; Troost, M.; Faaij, A.; Turkenburg, W. A comparison of electricity and hydrogen production systems with CO_2 capture and storage. Part A: Review and selection of promising conversion and capture technologies. *Prog. Energy Combust. Sci.* **2006**, *32*, 215−246.

(4) Ziems, C.; Meinke, S.; Nocke, J.; Weber, H.; Hassel, E. *Auswirkungen von fluktuierender Windenergieeinspeisung auf das regel- und thermodynamische Betriebsverhalten konventioneller Kraftwerke in Deutschland, Teil II—Auswirkungen großer Windeinspeisungen auf den zukünftigen Kraftwerkspark und dessen Tageseins (Effects of fluctuating Wind Power Feed to the Rule- And Thermodynamic Performance of Conventional Power Plants in Germany, Part II—Effects of Large Wind Feeds on the Future Power Plant and Its Day One)*; Projektnummer 333; VGB PowerTech e.V.: Essen, Germany, 2012.

(5) Saint-Drenan, Y.-M.; von Oehsen, A.; Gerhardt, N.; Sterner, M.; Bofinger, S.; Rohrig, K. *Dynamische Simulation der Stromversorgung in Deutschland nach dem Ausbauszenario der Erneuerbaren-Energien-Branche (Dynamic Simulation of Power in Germany after the Expansion Scenario in the Renewable Energies Industry)*; Fraunhofer Institut für Windenergie und Energiesystemtechnik (IWES): Bremerhaven, Kassel, Germany, 2009.

(6) Casella, F.; Colonna, P. Dynamic modeling of IGCC power plants. *Appl. Therm. Eng.* **2012**, *35*, 91−111.

(7) Damen, K.; Gnutek, R.; Kaptein, J.; Nannan, N. R.; Oyarzun, B.; Trapp, C.; Colonna, P.; van Dijk, E.; Gross, J.; Bardow, A. Developments in the pre-combustion CO_2 capture pilot plant at the Buggenum IGCC. *Energy Procedia* **2011**, *4*, 1214−1221.

(8) Robinson, P.; Luyben, W. Integrated gasification combined cycle dynamic model: H_2S absorption/stripping, water-gas shift reactors, and CO_2 absorption/stripping. *Ind. Eng. Chem. Res.* **2010**, *49*, 4766−4781.

(9) Bhattacharyya, D.; Turton, R.; Zitney, S. Control system design for maintaining CO_2 capture in IGCC power plants while load-following. Proceedings of the 29th Annual International Pittsburgh Coal Conference, Pittsburgh, United States, October 15−18. 2012; pp 2160−2173.

(10) Zitney, S.; Liese, E.; Mahapatra, P.; Turton, R.; Bhattacharyya, D.; Provost, G. AVESTAR Center: Dynamic simulation-based collaboration toward achieving operational excellence for IGCC plants with carbon capture. Proceedings of the 29th Annual International Pittsburgh Coal Conference, Pittsburgh, United States, October 15−18. 2012; pp 2093−2147.

(11) Schoen, P. Dynamic modeling and control of integrated coal gasification combined cycle units. Ph.D. Thesis, Delft University of Technology, 1993.

(12) Robinson, P.; Luyben, W. Plantwide control of a hybrid integrated gasification combined cycle/methanol plant. *Ind. Eng. Chem. Res.* **2011**, *50*, 4579−4594.

(13) Choi, Y.; Li, X.; Park, T.; Kim, J.; Lee, J. Numerical study on the coal gasification characteristics in an entrained flow coal gasifier. *Fuel* **2001**, *80*, 2193−2201.

(14) Chen, C.; Horio, M.; Kojima, T. Numerical simulation of entrained flow coal gasifiers. Part I: Modeling of coal gasification in an entrained flow gasifier. *Chem. Eng. Sci.* **2000**, *55*, 3861−3874.

(15) Chen, C.; Horio, M.; Kojima, T. Numerical simulation of entrained flow coal gasifiers. Part II: Effects of operating conditions on gasifier performance. *Chem. Eng. Sci.* **2000**, *55*, 3875−3883.

(16) Watanabe, H.; Otaka, M. Numerical simulation of coal gasification in entrained flow coal gasifier. *Fuel* **2006**, *85*, 1935−1943.

(17) Robinson, P.; Luyben, W. Simple dynamic gasifier model that runs in aspen dynamics. *Ind. Eng. Chem. Res.* **2008**, *47*, 7784−7792.

(18) Kasule, J.; Turton, R.; Bhattacharyya, D.; Zitney, S. Mathematical modeling of a single-stage, downward-firing, entrained-flow gasifier. *Ind. Eng. Chem. Res.* **2012**, *51*, 6429−6440.

(19) Seliger, B.; Hanke-Rauschenbach, R.; Hannemann, F.; Sundmacher, K. Modelling and dynamics of an air separation rectification column as part of an IGCC power plant. *Sep. Purif. Technol.* **2006**, *49*, 136−148.

(20) Mahapatra, P.; Bequette, B. Design and control of an elevated-pressure air separations unit for IGCC power plants in a process simulator environment. *Ind. Eng. Chem. Res.* **2013**, *52*, 3178−3191.

(21) Koch, I.; Hannemann, F.; Hoffmann, U. Dynamic simulation of operating cases and malfunctions of an IGCC power plant fuel system. *Chem. Eng. Technol.* **1999**, *22*, 568−570.

(22) Heil, S.; Brunhuber, C.; Link, K.; Kittel, J.; Meyer, B. Dynamic Modelling of CO_2-removal units for an IGCC power plant. Proceedings of the 7th Modelica Conference, Como, Italy, Sep. 20−22. 2009; pp 77 − 85.

(23) J-Power, The Future of Coal-Fired Thermal Power Generation. www.jpower.co.jp, (accessed 2011).

(24) Casero, P.; Peña, F.; Coca, P.; Trujillo, J. ELCOGAS 14 MWth pre-combustion carbon dioxide capture pilot. Technical & economical achievements. *Fuel* **2014**, *116*, 804−811.

(25) Wang, M.; Lawal, A.; Stephenson, P.; Sidders, J.; Ramshaw, C. Post-combustion CO_2 capture with chemical absorption: A state-of-the-art review. *Chem. Eng. Res. Des.* **2011**, *89*, 1609−1624.

(26) Kvamsdal, H.; Chikukwa, A.; Hillestad, M.; Zakeri, A.; Einbu, A. A comparison of different parameter correlation models and the validation of an MEA-based absorber model. Energy Procedia. *Energy Procedia* **2011**, *4*, 1526−1533.

(27) Åkesson, J.; Faber, R.; Laird, C.; Tummescheit, H.; Velut, S.; Zhu, Y. Models of a post-combustion absorption unit for simulation,

optimization and non-linear model predictive control schemes. Proceedings of the 8th International Modelica Conference, Dresden, Germany; 2011.
(28) Biliyok, C.; Lawal, A.; Wang, M.; Seibert, F. Dynamic modelling, validation and analysis of post-combustion chemical absorption CO_2 capture plant. *Int. J. Greenhouse Gas Control* **2012**, *9*, 428−445.
(29) Martelli, E.; Kreutz, T.; Consonni, S. Comparison of coal IGCC with and without CO_2 capture and storage: Shell gasification with standard vs partial water quench. *Energy Procedia* **2009**, *1*, 607−614.
(30) Carbo, M.; Boon, J.; Jansen, D.; van Dijk, H.; Dijkstra, J.; van den Brink, R.; Verkooijen, A. Steam demand reduction of water-gas shift reaction in IGCC power plants with pre-combustion CO_2 capture. *Int. J. Greenhouse Gas Control* **2009**, *3*, 712−719.
(31) Mattsson, S.; Elmqvist, H.; Otter, M. Physical system modeling with Modelica. *Control Eng. Practice* **1998**, *6*, 501−510.
(32) Fritzson, P. *Principles of Object-Oriented Modeling and Simulation with Modelica 3.3: A Cyber-Physical Approach*; Wiley-IEEE Press: Hoboken, NJ, in press.
(33) Gross, J.; Sadowski, G. Perturbed-Chain SAFT: An equation of state based on a perturbation theory for chain molecules. *Ind. Eng. Chem. Res.* **2001**, *40*, 1244−1260.
(34) Nannan, N. R.; de Servi, C. M.; van der Stelt, T.; Colonna, P.; Bardow, A. An equation of state based on PC-SAFT for physical solvents composed of polyethylene glycol dimethylethers. *Ind. Eng. Chem. Res.* **2013**, *52*, 18401−18412.
(35) Colonna, P.; van der Stelt, T.; Guardone, A. *FluidProp: A Program for the Estimation of Thermophysical Properties of Fluids*, version 3.0; Asimptote: Delft, The Netherlands, 2004−2012; http://www.FluidProp.com.
(36) Trapp, C.; Casella, F.; van der Stelt, T. P.; Colonna, P. Use of External Fluid Property Code in Modelica for Modelling of a Pre-combustion CO_2 Capture Process Involving Multi-Component, Two-Phase Fluids. Proceedings of the 10th Modelica Conference, Lund, Sweden, March 10−12. 2014.
(37) Casella, F.; Leva, A. Modelling of Thermo-hydraulic power generation processes using Modelica. *Math. Comput. Model. Dyn. Syst.* **2006**, *12* (1), 19−33.
(38) Casella, F.; Leva, A. Object-oriented modelling and simulation of power plants with modelica. Proceedings of the 44th IEEE Conference on Decision and Control, and the European Control Conference, CDC-ECC '05. 2005; pp 7597−7602.
(39) Trapp, C. *Development and validation of dynamic models of the shifting section of the CO_2 Catch-up pilot plant*; Doc. No. CO2 2013/0024, Delft University of Technology, Kluyverweg 1, 2629 HS Delft, 2013.
(40) Trapp, C. Advances in Model-Based Design of Flexible and Prompt Energy Systems - The CO_2 Capture Plant at the Buggenum IGCC Power Station as a Test Case. Ph.D. Thesis, Delft University of Technology, 2014.
(41) Seader, J. D.; Henley, E. J. *Separation Process Principles*; Wiley: Hoboken, NJ, 1998.
(42) Stichlmair, J.; Bravo, J. L.; Fair, J. General model for prediction of pressure drop and capacity of countercurrent gas/liquid packed columns. *Gas Sep. Purif.* **1989**, *3*, 19−28.
(43) Billet, R.; Schultes, M. Prediction of mass transfer columns with dumped and arranged packings: updated summary of the calculation method of billet and schultes. *Chem. Eng. Res. Des.* **1999**, *77* (Issue 6), 498−504.
(44) Trapp, C.; de Servi, C.; Casella, F.; Bardow, A.; Colonna, P. Dynamic modelling and validation of pre-combustion CO_2 absorption based on a pilot plant at the Buggenum IGCC power station. *Int. J. Greenhouse Gas Control* **2014**, submitted for publication.
(45) Luyben, W. L. *Process Modeling, Simulation, And Control for Chemical Engineers*; McGraw-Hill: New York, 1990.