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## Semi-empirical model for the direct simulation of power plant with integrated post-combustion CO<sub>2</sub> capture processes by wet chemical absorption

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### Abstract

In this work, a semi-empirical column model is developed to represent post-combustion CO<sub>2</sub> capture processes with chemical solvents in coal-fired steam power plants. The solvents are represented by empirical correlations on the basis of fundamental measurement data (CO<sub>2</sub> solubility, heat capacity, density). The model of a CO<sub>2</sub> capture process including the column model is coupled to detailed models of a hard-coal-fired steam power plant and of a CO<sub>2</sub> compressor to evaluate and compare the impact of CO<sub>2</sub> capture on the overall power plant process using six different solvents.

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Keywords: Energetic evaluation; Solvents; Monoethanolamine; Piperazine; Aminomethylpropanol; Potassium; Methyl-diethanolamine;

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### 1. Introduction

The performance of state-of-the-art steam power plants and the impact of optimisation measures can be evaluated with a high degree of accuracy by using modern simulation tools that have been developed for this particular purpose. Post-combustion CO<sub>2</sub> capture (PCC) by chemical absorption can be modelled with the help of equally powerful simulation tools developed for chemical and process engineering purposes.

The application of a PCC process in a coal-fired steam power plant is associated to a considerable net efficiency penalty of 8–12%-pts. Besides considerable capital expenditures for engineering, procurement, and construction of a PCC process, the loss in power output leads to significant costs for the operation of a coal-fired steam power plant with CO<sub>2</sub> capture. Thus, optimisation of the overall process and reduction of the efficiency penalty are required for a commercial deployment of this technology and an application at industrial scale. An integrated model of the overall process would allow evaluating the impact of the choice of chemical solvent, process configuration, and operational parameters on the performance, i.e. the net power output, of the power plant.

However, accurate models of steam power plants as well as of CO<sub>2</sub> capture processes are complex. Consequently, simulations are computationally intensive and a direct link between the two models is intricate. Such connection is hence either infeasible or does not allow to consider all key process parameters in the optimisation of the overall process with reasonable effort and within a reasonable timeframe.

There exists a variety of studies that focus on the optimal integration of PCC processes into coal-fired steam power plants (e. g. [1, 2]). Many of these studies, however, use simulation tools which are not specifically designed to represent the complex interrelations in power plant processes and suffer from a high degree of simplification.

A chemical absorption process for CO<sub>2</sub> capture based on an aqueous solution of monoethanolamine (MEA) is often considered as the benchmark since commercial processes with MEA which are designed for the application with coal-derived flue gas already exist. Consequently, the majority of scientific publications in the area of CO<sub>2</sub> capture focuses on MEA. Information on novel solvents that have been identified as promising candidates for CO<sub>2</sub> capture from flue gas, however, is scarce and usually focuses on more fundamental research aspects than on the energetic performance of the capture unit or the overall process.

The aim of this work is the energetic evaluation and comparison of chemical solvents for post-combustion CO<sub>2</sub> capture in a typical absorption-desorption process integrated into a coal-fired steam power plant. The performance of the overall process depends on the individual solvent properties in combination with the choice of process parameters of the CO<sub>2</sub> capture unit. A fair comparison on the basis of constant boundary conditions and assumptions therefore requires the optimisation of the process parameters for each solvent with respect to a minimal impact on the power plant.

To accomplish this aim, the overall process – including the power plant, the CO<sub>2</sub> capture unit and the CO<sub>2</sub> compressor – must be represented in adequate detail. The process model should be capable of representing any novel chemical solvent for which a minimal amount of information and measurement data is available. To account for the intricate interaction of the CCU with the steam power process and to allow for comprehensive sensitivity analyses of the key process parameters, the overall process model needs to be developed in a single simulation environment. To ensure the practicability and relevance of the overall process model, the use of a simulation tool which has proven its applicability in the power generation industry is favourable.

## 2. Methodology

EBSILON<sup>®</sup>*Professional* complies with the above list of requirements [3]. It is a simulation tool that is widely used for planning, construction, optimisation and control of thermal power plants and its solution algorithm has proven to be fast and to show favourable convergence properties to solve the mass- and energy balances of power plant processes. It comprises detailed models of standard components such as heat exchangers, pumps and compressors. Provided that solvent properties such as density and heat capacity are made available in adequate form, the simulation tool is therefore capable of representing most of the components in a CO<sub>2</sub> capture process. Absorption and desorption columns, however, cannot be modelled with EBSILON<sup>®</sup>, as it is not capable of representing the thermodynamic coherences which determine the transition of a gaseous species into a chemical solvent or vice versa.

The equilibrium of a gas mixture and a chemical solvent in aqueous solution as a function of pressure and temperature is commonly represented by complex rigorous thermodynamic models that consist of a large number of non-linear equations. This system of equations would have to be solved in parallel to the equally complex equation system that represents mass and energy balances of the power plant process. This task is impeded by the considerable computational effort related to the solution of these coupled systems and the lack of rigorous thermodynamic models for novel solvents for which only limited data is available. Therefore, a simplified representation of solvent properties with regard to the development of an accurate column model is necessary.

In the semi-empirical column model that is developed in this work, CO<sub>2</sub> solubility and other solvent properties are represented by employing empirical correlations that describe the relations between the measured parameters in a suitable mathematical manner. Although physical dependencies of these parameters are not taken into account, the approach allows a continuous representation of the H<sub>2</sub>O-CO<sub>2</sub>-solvent system in the range of temperatures and CO<sub>2</sub> partial pressures relevant for CO<sub>2</sub> capture processes in coal-fired steam power plants. The developed column model can be used with any chemical solvent for which a minimal amount of information is available.

Due to the large diversity of steam power plant types, chemical solvents, and possible capture process configurations, the outlined aim of this work is pursued by employing the example of a state-of-the-art hard-coal-fired steam power plant in combination with a simple, yet typical, absorption-desorption-type CO<sub>2</sub> capture process using the six chemical solvents listed in Table 1.

Table 1: Solvents selected for energetic evaluation

long name	abbreviation	composition		type
		mol / kg H <sub>2</sub> O (molality)	wt.-%	
monoethanolamine	MEA	7.0	30	primary amine
piperazine	PZ	8.0	41	polyamine
aminomethylpropanol	AMP	4.8	30	sterically hindered amine
potassium (carbonate)	K/PZ	5.0 / 2.5	14 / 15	(promoted) alkali salt solution
methyldiethanolamine	MDEA/PZ	7.0 / 2.0	42 / 8.6	amine blend
	AMP/PZ	5.0 / 2.5	27 / 13	amine blend

### 2.1. Power plant

The impact of a CO<sub>2</sub> capture process is evaluated using a hard-coal fired power plant with a gross power output of 1137 MW<sub>el</sub>. A bituminous coal with a lower heating value of 25.0 MJ/kg and specific CO<sub>2</sub> emissions of 343.6 g/kWh<sub>th</sub> is considered. The main features and parameters of this power plant at design conditions (full load without CO<sub>2</sub> capture) are given in Table 2. The flue gas parameters are given in Table 3.

The heat to desorb the CO<sub>2</sub> from the solution in the desorber column of the CO<sub>2</sub> capture unit is provided by extracting steam from the IP/LP crossover in the power plant. For a detailed explanation of the integration principles, refer to [4].

Table 2: Key parameters of power plant model

heat input	2306 MW <sub>th</sub>
gross power output	1137 MW <sub>el</sub>
net power output	1051 MW <sub>el</sub>
gross efficiency (LHV)	49.3 %
net efficiency (LHV)	45.6 %
live steam mass flow	826.5 kg/s
live steam pressure	285 bar
live steam temperature	600 °C
reheat pressure	59 bar
reheat temperature	620 °C
boiler feed water temperature	306.9 °C
condenser pressure	49/39 mbar
IP/LP crossover pressure	3.9 bar
feed water tank pressure	11.2 bar
cooling water temperature	19 °C
cooling water temperature gain	11 K

Table 3: Flue gas of reference steam power plant

temperature	48.5 °C
pressure	1.019 bar
mass flow	1054 kg/s
volume flow	942.3 m <sup>3</sup> /s
<i>composition</i>	
CO <sub>2</sub>	13.9 vol.-%
N <sub>2</sub>	70.7 vol.-%
H <sub>2</sub> O	11.2 vol.-%
O <sub>2</sub>	3.3 vol.-%
Ar, SO <sub>x</sub> , NO <sub>x</sub>	0.9 vol.-%

### 2.2. CO<sub>2</sub> compressor

To transport the separated CO<sub>2</sub> to an adequate storage site, a pipeline pressure of 110 bar is assumed to be sufficient. As the CO<sub>2</sub> leaving the overhead condenser downstream of the desorber at 40 °C is water saturated, the compressor has to cope with moist CO<sub>2</sub>. In this work an integrally-gearred (radial) compressor with six stages is considered. The auxiliary power duty and cooling duty of the CO<sub>2</sub> compressor are calculated using two empirical correlations given by LIEBENTHAL *et al.* [4].

### 2.3. CO<sub>2</sub> capture unit

The flow sheet of a standard absorption-desorption type CO<sub>2</sub> capture unit (CCU) is set up in EBSILON<sup>®</sup> (cf. Figure 1). The boundary conditions of the CCU are shown in Table 4. Most components of the CCU can be

represented by the standard components provided by the simulation tool. For the representation of the absorber and desorber column, the semi-empirical column model as explained in the following section is implemented.

Table 4: Boundary conditions of CO<sub>2</sub> capture unit

Temp. difference in rich-lean HX	5 K
reboiler temperature approach	10 K
CO <sub>2</sub> capture rate	90 %
absorber solution inlet temperature	40 °C
absorber flue gas inlet temperature	40 °C
flue gas cooler pressure drop	10 mbar
absorber pressure drop	50 mbar
desorber pressure drop	30 mbar
pump efficiency	80 %
fan isentropic efficiency	75 %
fan mechanical efficiency	95 %

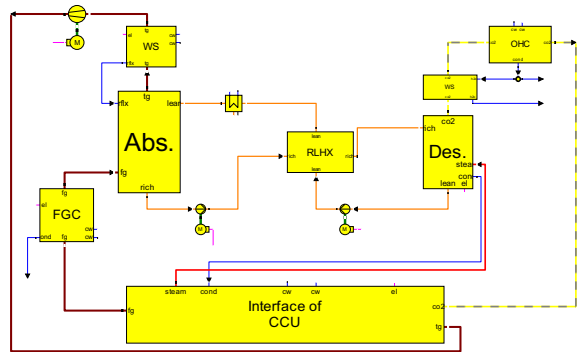


Figure 1: CO<sub>2</sub> capture unit in EBSILON® Professional

### 2.4. Semi-empirical column model

The semi-empirical column model in this work is based on a desorber model by OYENEKAN [143] but is modified and extended in the following points:

- To ensure an adequate representation of the absorber, nitrogen is considered as third gas component.
- The representation of the partial pressure of water takes into account the mole fraction of water in solution.
- The heat capacity of the selected solvents is represented as a function of temperature and CO<sub>2</sub> loading.
- Heat of vaporisation of water is considered as a function of temperature.
- Correlations for the representation of the vapour-liquid-equilibrium of the system CO<sub>2</sub>-H<sub>2</sub>O-solvent are modified and extended to the six solvents selected for evaluation in this work.

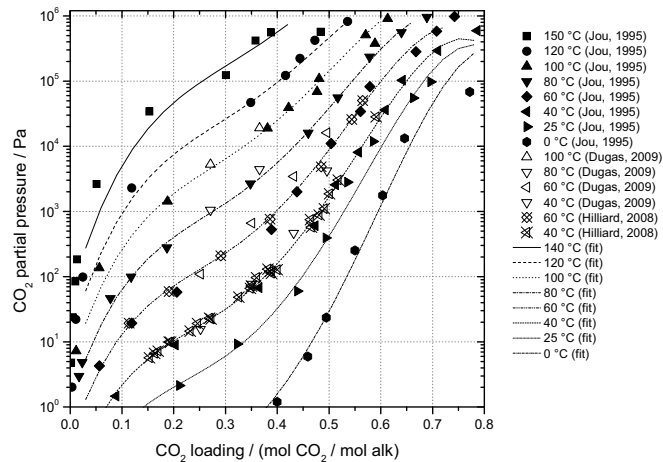
The column model consists of a series of equilibrium stages, where the MESH-equations (mass balance, equilibrium, summation, enthalpy balance) are setup for each stage. In equilibrium stage models it is assumed that the gas and liquid phase leaving the stage are in temperature and phase equilibrium. Evaporation of solvent is neglected; therefore, the gas phase consists of H<sub>2</sub>O, CO<sub>2</sub>, and – in case of the absorber – N<sub>2</sub> only.

The solubility of CO<sub>2</sub> in the solution is represented by empirical correlations which equate the equilibrium partial pressure of CO<sub>2</sub> in the gas phase with temperature and CO<sub>2</sub> loading of the liquid phase. Figure 2 shows the CO<sub>2</sub> solubility data of a 7 m MEA solution and the characteristics of the fitted correlations. The heat of absorption is derived from the CO<sub>2</sub> solubility data via the Gibbs-Helmholtz equation [5]. Additional correlations are derived for the representation of heat capacity and density of the CO<sub>2</sub>-loaded solution. Table 5 shows the underlying measurement data that were used in the derivation of the correlations.

The semi-empirical column model is validated by measurement data of a pilot plant operated at Esbjerg power station which was operated with 7 m MEA [6, 7].

Table 5: Used measurement data for derivation of correlations representing CO<sub>2</sub> solubility, heat capacity and density of selected solvents

solvent	CO <sub>2</sub> solubility	heat capacity	density
MEA	[8, 9, 10]	[11, 9]	[12, 13]
PZ	[14, 9, 10]	[9, 15]	[15]
AMP	[16, 17]	[18]	[19]
K/PZ	[20, 9]	[9]	[20]
MDEA/PZ	[21, 22]	[23]	[23]
AMP/PZ	[24]	[25]	[26]

Figure 2: CO<sub>2</sub> solubility of 7 m MEA

#### 2.4.1. Desorber

For solvents that show mass transfer coefficients for CO<sub>2</sub> comparable to MEA it can be assumed that equilibrium is reached in the desorber column of the CO<sub>2</sub> capture process (“equilibrium pinch”, q. v. [27]). It shows that an equilibrium stage model with 20 stages adequately describes the approach to equilibrium conditions in the desorber.

#### 2.4.2. Absorber

The aim of an optimal absorber design is to increase the amount of CO<sub>2</sub> that is absorbed by the solution, i. e. to achieve a CO<sub>2</sub> loading of the rich solution which is as high as possible. The higher this quantity, the lower the required solution circulation rate and consequently the lower the reboiler heat duty. As the temperature and concentration profile over the absorber column height is rate-determined, phase equilibrium between the gas and liquid outlet streams of the individual stages can no longer be assumed (Murphree efficiency ≠ 1). Absorber columns therefore commonly do not suffer from an equilibrium pinch, thus from a lack in driving force. The rate of absorption is rather limited by a rate pinch, which is due to the lower temperature and the low concentration of free alkali species. The rich loading is therefore determined by the temperature profile (impact on equilibrium loading) and the column design (impact on mass transfer rate, i. e. on approach to equilibrium). Hence, to predict the performance of an absorber, a rate-based model is necessary. As the development of detailed rate-based models for all of the six selected solvents is out of the scope of this work, a simplified approach for the representation of the absorber is employed.

The flue gas enters the absorber with a temperature of 40 °C and a CO<sub>2</sub> partial pressure of approx. 15 kPa. If equilibrium was reached at the absorber inlet then, depending on the solvent, a certain CO<sub>2</sub> loading of the rich solution corresponding to an equilibrium CO<sub>2</sub> partial pressure of 15 kPa could be achieved. However, a minimal driving force for CO<sub>2</sub> absorption is required. With a CO<sub>2</sub> partial pressure of 15 kPa in the flue gas, an equilibrium CO<sub>2</sub> partial pressure of 5 kPa allows for a partial pressure ratio of 3. This provides for a reasonable driving force which is achievable with economically feasible packing heights in the absorber [10]. Although the flue gas enters the absorber at a temperature of 40 °C, the corresponding loadings of the rich solution are calculated at 50 °C to account for the temperature increase which is related to the exothermic reactions in the absorber.

#### 2.5. Overall process model in EBSILON<sup>®</sup> Professional

Trilinos was identified as a suitable library of algorithms to solve the non-linear equation system that represents the semi-empirical column model of absorber and desorber in the overall process model. Trilinos provides fast and robust algorithms and since it is implementable in C++ it is compatible to the Programmable DLL (ProgDll, component 65) in EBSILON<sup>®</sup> [28, 29]. The solver is implemented together with routines for the setup of the

equation systems and the corresponding Jacobi matrix, for the control of input and output variables, and for the initialisation of the variables in an external direct link library (DLL). The DLL is called by the ESBILON<sup>®</sup> kernel via the two ProgDll components which represent the absorber and desorber column in the overall process model.

### 3. Results

The retrofit of a CO<sub>2</sub> capture process to a coal-fired steam power plant leads to a reduction in net power output due to the required steam extraction for the regeneration of the solution in the desorber (50–70 % of the overall net efficiency penalty), due to the auxiliary power demand for CO<sub>2</sub> compression (20–40 %), and due to the auxiliary power of fans as well as circulation and cooling water pumps in CCU and CO<sub>2</sub> compressor.

The solvent and the chosen process parameters of the CCU thereby influence the magnitude of the reduction in power output and net efficiency of the power plant. Two of the most critical process parameters in the CCU are the  $L/G$ <sup>1</sup> in the absorber and the reboiler temperature in the desorber. Figure 3 shows the reboiler heat duty of the CCU for the six evaluated solvents. The  $L/G$  and the reboiler temperature are varied, whereas all other process parameters are kept constant (q. v. Table 4).

The occurrence of a minimal reboiler heat duty for a certain  $L/G$  is due to two opposing effects: when the amount of circulated solution is reduced, less sensible heat is needed to increase the temperature of the solution from the level at the desorber inlet to the reboiler temperature; however, a lower solution flow rate requires a lower CO<sub>2</sub>-loading of the lean solution and correspondingly a higher CO<sub>2</sub> capacity to achieve a specified CO<sub>2</sub> capture rate. To achieve a lower lean loading, more stripping steam is needed. Therefore, more heat is required for the evaporation of water when the  $L/G$  is reduced<sup>2</sup>. Note that the increase in solution flow rate is limited, as a minimal difference in rich and lean loading is needed to maintain a minimal capacity for CO<sub>2</sub> uptake in the absorber.

Figure 4 shows the overall net efficiency penalty for the six solvents compared to the reference power plant without CO<sub>2</sub> capture. The impact of the reboiler temperature on the overall net efficiency is mainly determined by three effects:

1. All six evaluated solvents have a high heat of absorption of CO<sub>2</sub> and therefore show a decreasing reboiler heat duty when the reboiler temperature is increased<sup>3</sup>.
2. Neglecting solvent evaporation, the total pressure in the desorber is simply the sum of partial pressures of CO<sub>2</sub> and H<sub>2</sub>O:

$$p_{\text{des}} = p_{\text{CO}_2}(T_{\text{reb}}, \alpha_{\text{lean}}) + p_{\text{H}_2\text{O}}(T_{\text{reb}}).$$

To achieve a specified CO<sub>2</sub> capture rate in the absorber, the circulated solution needs to be regenerated to a certain lean CO<sub>2</sub> loading  $\alpha_{\text{lean}}$  in the desorber. An increase in reboiler temperature  $T_{\text{reb}}$  while keeping a constant lean CO<sub>2</sub> loading leads to an increase in both the partial pressure of CO<sub>2</sub> ( $p_{\text{CO}_2}$ ), due to the characteristics of CO<sub>2</sub> solubility, as well as in the partial pressure of H<sub>2</sub>O ( $p_{\text{H}_2\text{O}} = x_{\text{H}_2\text{O}} p_{\text{H}_2\text{O}}^s$ ), due to the elevated water vapour pressure  $p_{\text{H}_2\text{O}}^s$ . Therefore, an increase in reboiler temperature for  $\alpha_{\text{lean}} = \text{const.}$  requires an increase in desorber pressure  $p_{\text{des}}$  which in turn reduces the pressure ratio and the auxiliary power in the subsequent CO<sub>2</sub> compressor.

3. As the reboiler temperature determines the required quality of the steam which is extracted from the power plant, an increase of this quantity implies that steam at higher pressure is needed. The extraction of higher quality steam at a higher pressure results in larger specific power losses (e. g. in MW<sub>el</sub> per MW<sub>th</sub> extracted steam) [4].

An increase in reboiler temperature, limited by thermal degradation of the solvent, therefore has both positive (1. and 2.) and negative (3.) impacts on the net power output of the power plant. The overall net efficiency penalty therefore becomes minimal for a certain combination of  $L/G$  and reboiler temperature. Figure 4 shows for all six evaluated solvents that when the reboiler temperature is increased, the net efficiency penalty first decreases due to the positive effects of decreasing reboiler heat duty (1.) and decreasing auxiliary power for CO<sub>2</sub> compression (2.). The net efficiency penalty reaches a minimum for reboiler temperatures between 105 and 114 °C. If the reboiler temperature is further increased, the net efficiency penalty increases due to the increasing specific power losses related to the required steam extraction from the power plant at higher pressure (3.).

<sup>1</sup>  $L/G$  denotes the ratio of mass flows of liquid and gas in the column. In steady-state operation  $L/G$  is proportional to the solution flow rate.

<sup>2</sup> For a more detailed discussion of this subject refer, for example, to [27].

<sup>3</sup> For an explanation of this phenomenon, refer to [5].

The overall process evaluation which is based on fundamental measurement data (CO<sub>2</sub> solubility, heat capacity, and density) reveals that under the outline boundary conditions MDEA/PZ is best suited for a retrofit integration to the considered hard-coal-fired steam power plant. With this solvent the net efficiency penalty is 8.46 %-pts. compared to 10.15 %-pts. for MEA. PZ shows the lowest reboiler heat duty of 2.45 GJ<sub>th</sub> / t CO<sub>2</sub> at a reboiler temperature of 150 °C (MDEA/PZ: 2.52 GJ<sub>th</sub> / t CO<sub>2</sub> at 120 °C; MEA: 3.35 GJ<sub>th</sub> / t CO<sub>2</sub> at 120 °C). However, for the considered power plant, steam extraction at 6.9 bar as required to supply the heat to the reboiler at a temperature of 150 °C leads to approximately 46.6 % higher specific power losses than steam extraction at 3.0 bar which is required for a reboiler temperature of 120 °C (q. v. [4]). Therefore, PZ cannot benefit from the low reboiler heat duty at higher reboiler temperatures and shows a higher net efficiency penalty than MDEA/PZ for optimal process parameters in the evaluation of the overall process.

#### 4. Conclusions

A semi-empirical column model was developed and implemented in EBSILON<sup>®</sup>Professional which allows to represent a complete CO<sub>2</sub> capture process. The direct coupling of the CO<sub>2</sub> capture process model to detailed models of a hard-coal-fired steam power plant and of a CO<sub>2</sub> compressor allows the energetic evaluation and comparison of chemical solvents based on fundamental measurement data and on a fair and constant set of boundary conditions. In case of retrofit integration of a CO<sub>2</sub> capture process to the specific hard-coal fired power plant with a crossover pressure of 3.9 bar at design conditions (full load without CO<sub>2</sub> capture), the use of MDEA/PZ leads to the lowest overall net efficiency penalty of 8.46 %-pts.

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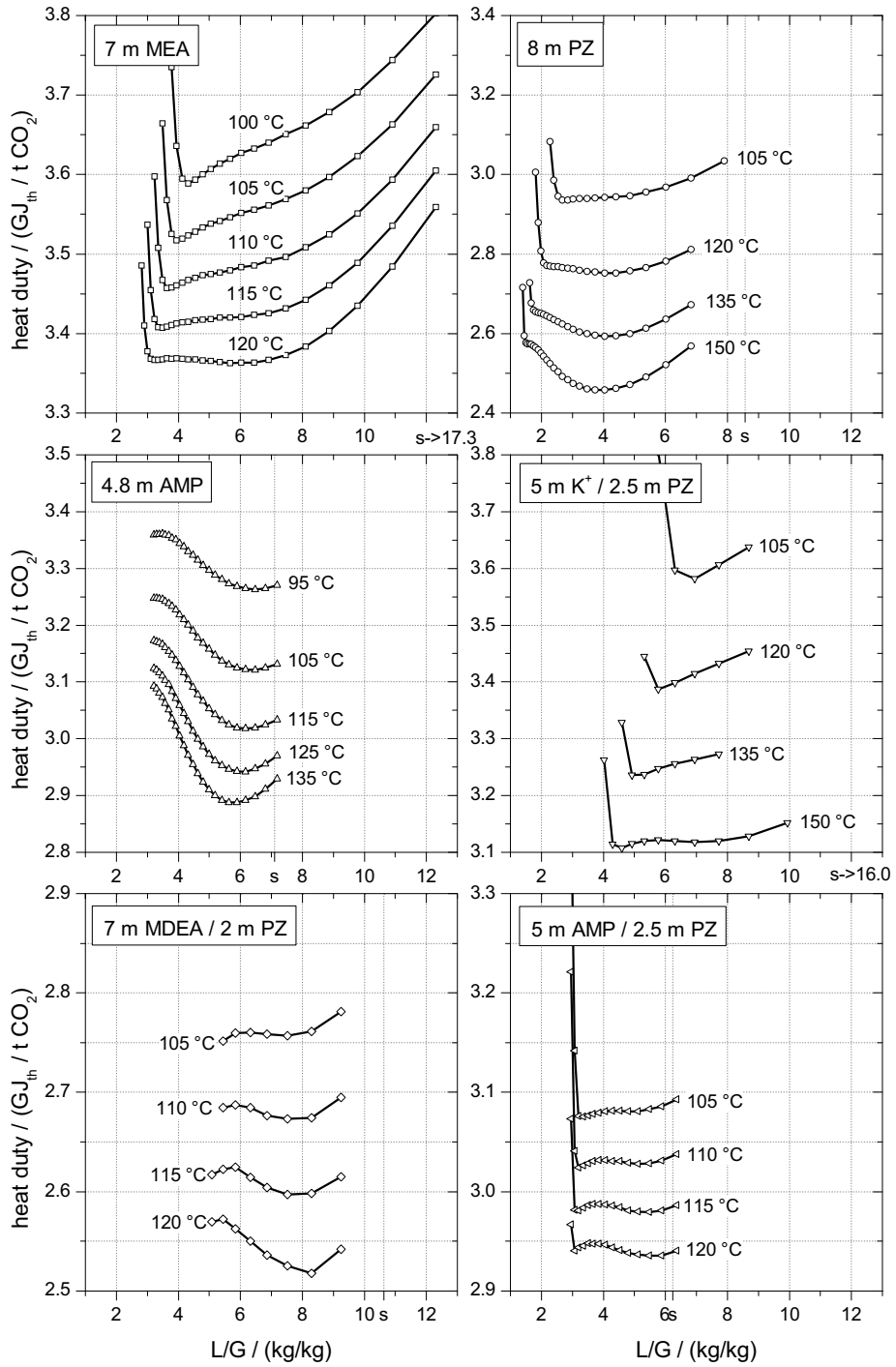


Figure 3: Reboiler heat duty for different reboiler temperatures and varying  $L/G$

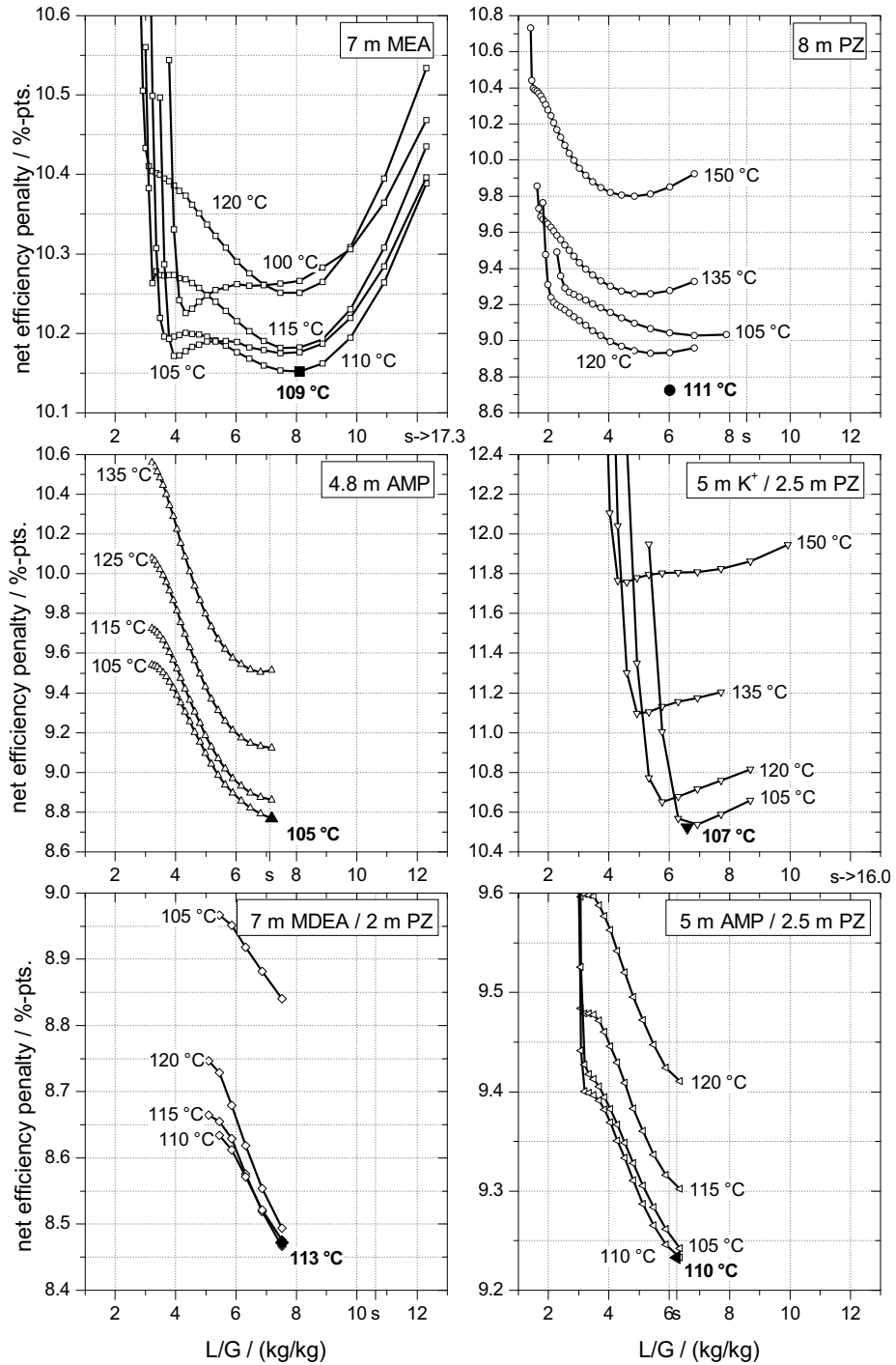


Figure 4: Net efficiency penalty in case of retrofit integration to reference power plant for different reboiler temperatures and varying L/G