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A Comparative Study of Carbon Capture for Different Power Plants

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Within the EU-funded project CAPSOL, capture units for three different power plants were designed. The solvents used were aqueous amine solutions; feed compositions and main process conditions were first determined using equilibrium-based simulations. As the main evaluation criterion, reaching a CO_2 absorption degree of 90 % with minimum energy requirements was selected. The equilibrium based simulations did not take real column design and internals into account. Therefore, a more rigorous, rate-based approach was used in our work to determine necessary column heights. It was shown that an absorption degree of 90 % is not always realizable and even not always required.

1. Introduction

Efficient carbon capture is currently regarded as one of mankind's greatest challenges, due to huge emissions threatening global warming. In 2012, according to the US Energy Information Administration (2015), 32,723 Mt CO_2 emerging from the use of fossil fuels were emitted worldwide and contributed to the environment pollution. About 40 % of these emissions stem from coal-fired power plants and about 20 % from the use of natural gas. Available capture technologies are predominantly based on the use of liquid solvents enhancing the separation efficiency by selectively dissolving CO_2, and optimisation studies of absorption-based capture processes are very intensive (Vaidya and Kenig, 2007).

One of the concerted actions in this area was the EU-funded collaborative project CAPSOL (FP7-ENERGY-2011-282789, full title “Design technologies for multi-scale innovation and integration in post-combustion CO_2 capture: From molecules to unit Operations and Integrated Plants”). In this project, several partners worked on different complementary topics, e.g. efficient solvent screening (Papadopoulos et al., 2014) and optimal flowsheet design (Damartzis et al., 2013). Our contribution was related to the theoretical and experimental analysis of absorption-desorption processes carried out in different power plants. In this paper, a comparative theoretical study of absorption columns for one gas-fired and two coal-fired power plants is presented.

A crucial evaluation criterion of CO_2 absorption processes is the energy demand of the total absorption-desorption loop, which is primarily determined by the solvent regeneration in the desorber. To address this issue, the data on the necessary heat duty for the total capture unit work were generated within CAPSOL based on simplified equilibrium-based models. However, such models are usually not sufficient for a comprehensive feasibility study with respect to the industrial realisation; instead, a more rigorous modelling framework has to be applied. We used the well-established rate-based approach implemented into the Aspen Custom Modeller software in order to determine column heights necessary to reach a CO_2 absorption degree of 90 %. These data were then delivered to another CAPSOL partner who carried out a complete column design study including packings, solvent redistributors, etc. This provides essential information for large-scale equipment design and feasibility analysis.

2. Process description

CO_2 capture from flue gases is typically done in an absorption-desorption loop. The raw gas stream is fed to the bottom of an absorption column and is sweetened by the counter-currently flowing solvent. Often,

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amine-based solvents are applied. The dissolved CO_2 is then removed from the solvent in the subsequent desorption/stripping column. The regenerated amine solution is recirculated to the top of the absorber. In order to reduce energy requirements, a heat exchanger is applied, in which the lean amine solution exiting the desorption column preheats the loaded amine solution entering the same column.

For this study, the state-of-the-art solvent monoethanolamine (MEA) was used. MEA, a primary amine, reacts with CO_2 to form carbamate species. The overall reaction proceeds through the formation of a zwitterion as an intermediate (Vaidya and Kenig, 2007). The whole reaction system involves a complex set of parallel and consecutive reactions in the liquid phase. Two of these reactions are reversible and kinetically controlled:

$$\mathrm{CO_2 + MEAH + H_2O \rightleftharpoons MEACOO^- + H_3O^+} \quad (1)$$
$$\mathrm{CO_2 + OH^- \rightleftharpoons HCO_3^-} \quad (2)$$

Three other reactions are reversible instantaneous:

$$\mathrm{MEAH + H_3O^+ \rightleftharpoons MEAH^+ + H_2O} \quad (3)$$
$$\mathrm{HCO_3^- + H_2O \rightleftharpoons CO_3^{2-} + H_3O^+} \quad (4)$$
$$\mathrm{2H_2O \rightleftharpoons H_3O^+ + OH^-.} \quad (5)$$

The reaction rate constants for Eq(1) and Eq(2) can be taken from Aspen Technology Inc. (2012) and the expressions for chemical equilibrium constants from Austgen (1989).

In the CAPSOL project, flue gases of three power plants based on different fuels, namely natural gas, bituminous coal and lignite, were considered. Data on these flue gases are summarized in Table 1. Along with CO_2, water (H_2O) and nitrogen (N_2), flue gases usually contain oxygen and argon; for the sake of simplicity, these components were treated in simulations cumulatively as N_2. The CO_2 concentrations for the coal cases are about 20 wt%, whereas for the gas case, it is only 7.5 wt%. The CAPSOL partners determined optimal values for the solvent specifications allowing reaching a CO_2 absorption degree (absorbed amount of CO_2 divided by the amount of CO_2 in the flue gas) of 90 % with minimum energy requirements. These data are based on equilibrium-stage simulations and also summarized in Table 1.

Table 1: Flue gas and solvent input parameters

	Natural gas-fired 420 MWe combined cycle gas turbine (gas case)	Bituminous coal-fired 420 MWe advanced supercritical power plant (bituminous-coal case)	Lignite-fired 650 MWe advanced supercritical power plant (lignite case)
	Flue gas		
p / bar	1.1	1.1	1.1
T / °C	54.28	41.93	51.6
$w(CO_2)$ / g/g	0.075	0.211	0.202
$w(H_2O)$ / g/g	0.057	0.043	0.074
$w(MEA)$ / g/g	0	0	0
$w(N_2)$ / g/g	0.867	0.746	0.723
$\dot{m}$ / kg/s	553.4	603.6	693.7
$\dot{V}$ / m^3/s	491.0	486	588.3
	Solvent		
T / °C	40	40	40
$\dot{m}$ / kg/s	900.5	2,426.2	2,273.7
$w(MEA)$ / g/g	0.27	0.305	0.301
$\frac{n(CO_2)}{n(MEA)}$ / mol/mol	0.26	0.27	0.23

These solvent specifications were determined without due regard to important design parameters, e.g. the necessary number of columns, column diameter, the packing type and height. For the present study, we focused on a standard packing with an interfacial area of 250 m^2/m^3 and an inclination angle of 45°. Among others, these values correspond to B1-250.45 packing by Julius Montz or to Sulzer Mellapak 250.Y. Considering the immense flue-gas volume flows and in order to keep the pressure drop low, the column diameter was set to 15 m. Furthermore, for the gas case, two columns were used in parallel, while for the coal-cases, three columns were used to split the huge flow.

3. Rigorous rate-based model

The model used in this work is a non-equilibrium stage model based on the description of a single stage representing a packing segment of a column. The gas and liquid phases are balanced separately, and mass- and heat-transfer resistances are considered according to the film theory, by directly accounting for the interfacial fluxes. The film model equations are combined with the relevant reaction and diffusion kinetics and include the specific features of electrolyte solutions. An important advantage of this model is that the flow peculiarities in the absorption column can be directly accounted for using correlations for hold-up, pressure drop, interfacial area and mass-transfer coefficients. The model permits the concentration and temperature profiles in the absorber to be calculated, both in film and bulk regions. The whole system of equations is solved simultaneously within the simulation tool Aspen Custom Modeller, and thus, the interactions between the film profiles and the bulk values as well as the mass transfer - reaction coupling are taken into account.

3.1 Model equations

In what follows, the most important equations are given; a complete model description can be found elsewhere (Kucka et al., 2003). The bulk-phase mass balance equations for both contacting phases consider the axial change of the total molar stream and the compositions. In Eq(6) a source term describing the chemical reaction is included:

$$\frac{\partial L x_i^{lb}}{\partial z} = n_i^{lb} a^i A_c + R_i^{lb} \phi^l A_c, \quad i = 1, \dots, n^l \tag{6}$$

$$\frac{\partial G y_i^{gb}}{\partial z} = n_i^{gb} a^i A_c, \quad i = 1, \dots, n^g. \tag{7}$$

The enthalpy balances for both phases are as follows:

$$\frac{\partial L h^{lb}}{\partial z} = q^{lf} a^i A_c \tag{8}$$

$$\frac{\partial G h^{gb}}{\partial z} = q^{gf} a^i A_c. \tag{9}$$

In the liquid film region, the interfacial fluxes of the components are calculated with the Nernst-Planck equation:

$$n_i^{lf} = -\frac{c_t^{lf} D_{eff,i}^l}{\delta^l} \left(\frac{\partial x_i^{lf}}{\partial \eta^l} + x_i^{lf} z_i \frac{F}{R_G T} \frac{\partial \varphi}{\partial \eta^l} \right) + x_i^{lf} n_t^{lf}, \quad i = 1, \dots, n^l. \tag{10}$$

As mass transfer and chemical reactions occur simultaneously, the component molar fluxes at the interface and at the boundary between the film and bulk phase are not equal. Therefore, a differential component balance has to be considered in the liquid film:

$$\frac{1}{\delta_l} \frac{\partial n_i^{lf}}{\partial \eta^l} - R_i^{lf} = 0, \quad i = 1, \dots, n^l. \tag{11}$$

In the gas film region, the component molar fluxes are described by

$$n_i^{gf} = -\frac{c_t^{gf} D_{eff,i}^g}{\delta^g} \frac{\partial y_i^{gf}}{\partial \eta^g} + y_i^{gf} n_t^{gf}, \quad i = 1, \dots, n^g. \tag{12}$$

The heat flux for both film regions is given by

$$q^{lf} = -\frac{\lambda^{lf}}{\delta^{lf}} \frac{\partial T^l}{\partial \eta^l} + \sum_{i=1}^{n^l} n_i^{lf} h_i^{lf} \tag{13}$$

$$q^{gf} = -\frac{\lambda^{gf}}{\delta^{gf}} \frac{\partial T^g}{\partial \eta^g} + \sum_{i=1}^{n^g} n_i^{gf} h_i^{gf} \tag{14}$$

At the phase interface, gas and liquid phase compositions are linked by the thermodynamic equilibrium condition:

$$y_i^i = K_{D,i} x_i^i, \quad i = 1, \dots, n^g.. \tag{15}$$

The film thicknesses δ^{lf} and δ^{gf} as well as the column hold-up ϕ are estimated via empirical mass transfer correlations (Billet and Schultes, 1999). For the effective interfacial area a^i, the correlation by Tsai et al. (2011) was used.

3.2 Model validation

The model was validated using experimental data by Notz et al. (2012). Experiments were performed in a pilot plant with an inner diameter of 0.125 m filled with 4.2 m height Mellapak 250.Y. Gas-phase temperature and liquid-phase CO_2 concentration profiles were measured along the column. Figure 1 shows a parity plot of our simulated absorption degrees and those measured by Notz et al (2012) for 10 experimental runs. The maximum deviation between the simulated and measured values was less than 10 %.

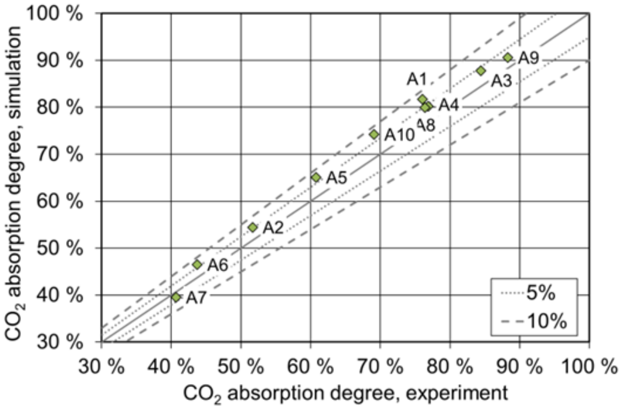

Figure 1: Parity plot of simulated compared to experimental CO_2 absorption degrees

4. Simulation studies

Simulation studies with the model described above were performed with the aim to determine the packing height necessary to reach a CO_2 absorption degree of 90 % for the flue-gas and solvent conditions given in Table 1.

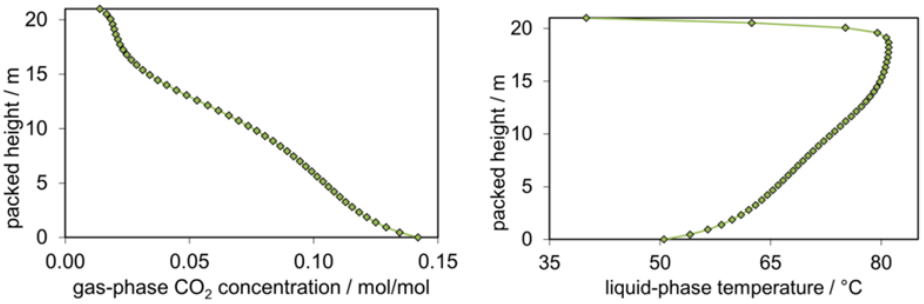

Figure 2: Gas-phase CO_2 concentration and liquid-phase temperature profile for the bituminous coal-fired case

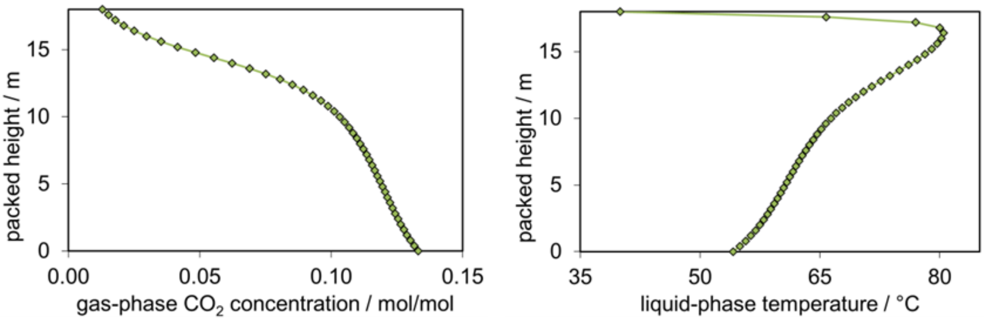

Figure 3: Gas-phase CO_2 concentration and liquid phase temperature profile for the lignite fired case

In case of the bituminous coal-fired power plant, the targeted absorption degree of 90 % could be reached with a packed height of 21 m. For the lignite-fired power plant, a packed height of 18 m is required. Figures 2 and 3 show the gas-phase CO_2 concentration profiles and the liquid-phase temperature profiles along the packed height. In both cases CO_2 is absorbed directly at the top of the column, where lean amine comes in contact with the gas. Due to exothermal reactions, the temperature increases very fast reaching values of about 80 °C. Consequently, absorption becomes slower and temperature decreases. The lower temperature leads again to a better absorption near the middle of the column.

In contrast to the coal cases, for the natural-gas-fired power plant, the absorption degree of 90 % could only be reached at packed heights above 60 m (Figure 4). Such columns can hardly be used industrially because of statical limitations. Furthermore, such a column would cause a very high pressure drop, which is disadvantageous when energy consumption should be reduced. For these reasons, it was decided to set the maximum packed height to 32 m.

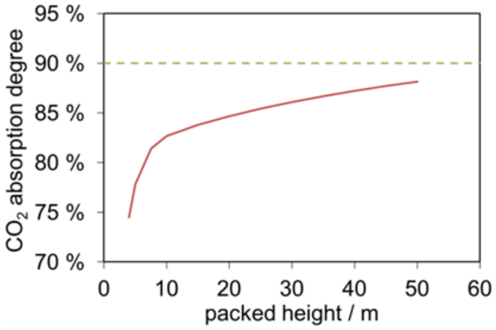

Figure 4: CO_2 absorption degree as a function of the packed height for the gas-fired case

Simulations performed for this packed height led to an absorption degree of 86 %. Although the value of 90 % could not be reached, the CO_2 concentration in the clean gas was around 600 ppm (Figure 5), which is close to the ambient air value and even lower than half of the concentration out of the coal cases (i.e., 1,390 ppm and 1,300 ppm).

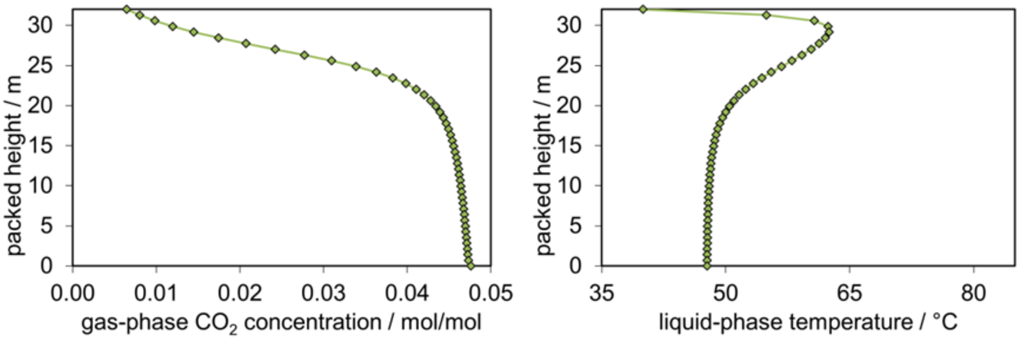

Figure 5: Gas-phase CO_2 concentration and liquid-phase temperature profile for the gas-fired case

Furthermore, Figure 5 shows that most of the CO_2 is absorbed in the upper part of the column. In the lower part, there is nearly no change in the concentration, because the solvent equilibrium loading is almost achieved. Consequently, the driving force for the CO_2 absorption drops and higher packed sections are needed.

5. Conclusions

In this paper, we presented simulation studies carried out within the EU-funded project CAPSOL. Rate-based simulations were performed in order to determine necessary heights of CO_2-absorption columns to be integrated into different power plants. It was shown, that in case of coal-fired power plants, an absorption degree of 90 % could be reached with industrially feasible column heights. In contrast, for the gas-fired power plant, such a degree could only be achieved with an unrealistically high column. Nevertheless, for a reduced absorption degree of 86 %, a fully feasible column height resulted, with a low CO_2 concentration of only 600 ppm in the outlet gas stream. These results demonstrate how important the determination of the column height for the feasible industrial design of capture plants is. Furthermore, it is illustrated that an extremely high absorption degree is not always meaningful.

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Notation

a	specific area, m^2/m^3
A_c	cross-section area of column, m^2
c_t	total molar concentration, mol/m^3
D_{eff}	effective diffusion coefficient, m^2/s
F	Faraday's constant (96,500 C/mol)
G	total gas molar flow, mol/s
K_D	phase distribution coefficient
L	total liquid molar flow, mol/s
n	number of components
n_i	molar flux of component i, mol/m^2s
p	pressure, bar
q	heat flux, J/m^2s
R	reaction rate, mol/m^3s
R_G	gas constant (8.3144 J/molK)
T	temperature, °C
w	mass fraction
x	liquid phase mole fraction
y	gas phase mole fraction
z	axial co-ordinate, m
z	electrical charge

Greek letters

δ	film thickness, m
η	dimensionless film co-ordinate
λ	thermal conductivity, W/mK
φ	electrical potential, V
ϕ	volumetric hold-up

Subscripts

i	component index

Superscripts

b	bulk
f	film
g	gas phase
i	interface
l	liquid phase

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