Process simulation of CO_2 absorption data fitted to performance efficiency at TCM Mongstad

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Abstract

This work's first aim is to fit the simulated results to pilot plant data from Test Centre Mongstad (TCM) for both high and low CO_2 exhaust gas inlet concentrations. The next aim is to evaluate whether fitted parameters for one scenario (a set of experimental data under specified conditions) give reasonable predictions under other conditions. The scenarios at TCM have been simulated in both a rate-based model in Aspen Plus (RateSep) and an equilibrium-based model in Aspen HYSYS. The rate-based model's performance data were fitted by only changing the liquid hold-up factor (multiplied with the hold-up estimated by the Bravo/Rocha/Fair correlations) to obtain the experimental CO_2 removal efficiency. In the equilibrium-based model, a Murphree efficiency (E_M) was specified for 24 or 18 stages (meter of packing) to fit the CO_2 removal efficiency and the temperature profile from performance data. The specified E_M profiles were then used to fit performance data for other scenarios by adjusting only an E_M-factor, multiplying all the E_M values in an E_M profile. The performance (CO_2 removal and temperature profile) was reasonably simulated for each given scenario for all the models. It is shown in this work that the use of the liquid hold-up factor (and not the interfacial area factor) is convenient to fit the rate-based model to the pilot plant data. Using fitted parameters at low CO_2 inlet concentration to predict performance at high concentration needed an adjustment factor (liquid hold-up factor or E_M-factor) to obtain correct CO_2 removal predictions. A liquid hold-up factor of 0.72 and an E_M-factor of 1.72 fitted to performance data for high CO_2 concentration at TCM gave reasonable predictions compared to performance data for high CO_2 concentration from the Esbjerg pilot plant.

Keywords: CO_2, amine, absorption, simulation

1. Introduction

The CO_2 Technology Centre Mongstad (TCM) is the world's largest test facility for CO_2 capture technology. To test CO_2 absorption into amine-based solvents, there is an absorption column with a rectangular cross-section equivalent to a packing diameter of 3 meters, and a packing height up to 24 meters. There have been performed performance tests of CO_2 absorption from flue gas into 30 wt.% monoethanolamine (MEA) in 2013 (Thimsen *et al*., 2014; Hamborg *et al*., 2014) and in 2015 (Gjernes *et al*., 2017; Faramarzi *et al*., 2017). Figure 1 shows the principle of the amine-based CO_2 absorption and the desorption facility at TCM. In this work, the evaluated parameters are especially the total CO_2 capture rate (in % of incoming CO_2) in the absorption section and the temperature profile from top to bottom.

In earlier work (Sætre, 2016; Øi *et al*., 2018; Fagerheim, 2019; Øi and Fagerheim, 2020), the equilibrium models (in Aspen Plus and Aspen HYSYS) were fitted to one specific scenario by adjusting the Murphree efficiency (E_M) for each stage, and the rate-based model (in Aspen Plus) was fitted to another scenario by changing the interfacial area factor. Razi *et al*. (2013) evaluated different correlations in rate-based simulations in Aspen Plus for predicting performance from the Esbjerg pilot plant. Some sets of correlations gave very good predictions, and some sets gave poor predictions of CO_2 removal and temperature profiles.

In rate-based models, it is standard to use the interfacial area factor to fit the model to performance data. In this work, the hold-up factor (multiplied with the hold-up estimated by in-built correlations) was used for the same purpose. One of the estimation methods in Aspen Plus for liquid hold-up is the Bravo/Rocha/Fair model (Rocha, 1992).

This work is based on the Master Thesis of Njål Sæter (2021) and also on earlier work at the University of South-Eastern Norway.

The first aim of this work is to compare results from simulations with performance data from TCM for CO_2 absorption into 30 wt.% MEA using both rate-based and equilibrium models. The second aim, which is specific to this work, is to test whether fitted parameters for one scenario (a set of experimental data at specified conditions) give reasonable predictions under other conditions. Especially it is evaluated what adjustment is necessary for using

fitted parameters for low CO_2 inlet concentrations to predict performance at high CO_2 inlet concentration.

2. Process simulation models for CO_2

Process simulation tools available for CO_2 absorption processes contain models for vapour/liquid equilibrium calculations and efficient solvers. The rate-based tools also include models for chemical, heat transfer and mass transfer kinetics. The commercially available programs Aspen Plus and Aspen HYSYS are used here. Both Aspen Plus and the new Aspen HYSYS acid gas model use an Electrolyte-NRTL equilibrium model based on Austgen *et al*. (1989). Other tools are described in Øi and Fagerheim (2020).

Aspen Plus has a rate-based model to describe the reactive absorption processes. The rate-based model is based on MERSHQ (material balances, energy balances, rate of mass and heat transfer, summation of composition, hydraulic equations for pressure drop and equilibrium) equations which are used to determine molar and energy fluxes transfer across the vapour-liquid interfaces.

The CO_2 capture plant at the University of Texas in Austin was modelled with ASPEN rates, a second-generation rate-based multistage separation unit operation model in ASPEN Plus. The parameters in this file are mainly based on the work of Zhang *et al*. (2009), who fitted Aspen Plus simulations to experimental runs at a CO_2 absorption pilot plant. Different rate-based models have been developed for TCM in the Master Thesis works of Desvignes (2015), Sætre (2016) and Fagerheim (2019).

Equilibrium based models assuming equilibrium at each stage can be extended by using a Murphree efficiency (the ratio of the change in mole fraction from a stage to the next divided by the change assuming equilibrium). An advantage of using Murphree efficiencies compared to rate-based simulations is that it is simpler and fewer parameters need to be specified. In the Master Thesis work of Zhu (2015) and Sætre (2016), a Murphree efficiency for each stage (meter of packing) was estimated for one set (scenario) of TCM data (Hamborg *et al*., 2014). A good agreement between the measured and simulated temperature profile was obtained using different fitted Murphree efficiencies for each stage. Luo *et al*. (2009) tested Aspen RadFrac, ProTreat, ProMax, Aspen RateSep, CHEMASIM from BASF and CO2SIM from SINTEF/NTNU by comparing with pilot plant data. The result was that all models could fit the CO_2 capture rate, but the temperature and concentration profiles were not well predicted. When comparing Aspen HYSYS and Aspen Plus, Øi (2012) claimed that there were only slight differences between the tested equilibrium models. A rate-based and equilibrium-based model with estimated Murphree efficiencies gave similar results. In the work by Øi *et al*. (2018), different models were compared for 4 scenarios from TCM at low CO_2 input concentrations (3-4 vol-%). 5 scenarios were compared. The results from these comparisons showed that the equilibrium and rate-based models performed equally well in both fitting performance data and in predicting performance at changed conditions.

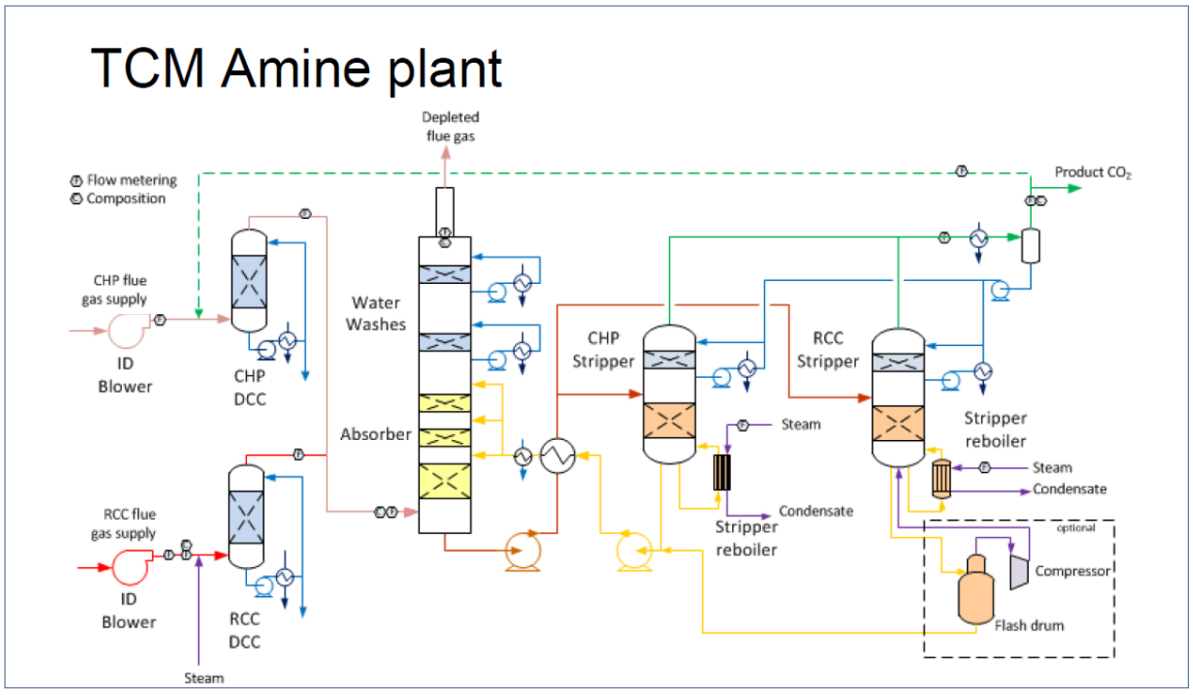

Figure 1: Simplified process diagram of the amine-based CO_2 capture plant at TCM (Hamborg *et al*., 2014)

3. Data and specifications

3.1. Performance data from TCM

Performance data with low CO_2 inlet concentration (3-4 vol-%) has been taken from 5 sets of conditions (scenarios) at TCM. They are from campaigns in 2013 and 2015 for approximately 30 wt.% MEA in water. 24 meters of packing height (the maximum available) were used in these scenarios.

The data (mainly conditions of the inlet gas stream and the inlet amine stream to the absorption section) for the 5 scenarios are listed in Tab. 1. The 5 scenarios which have been selected in this work are named H14 and 6w from 2013 (Hamborg *et al*., 2014), 2B5 and Goal1 from 2015 (Gjernes *et al*. 2017) and F17 from Faramarzi (2017). The names have been used internally at TCM, except for the H14 and F17 scenarios, which are named by first author's initial and year. The different scenarios cover typical conditions. 17F is at a low liquid to gas conditions, Goal1 and H14 are at low temperature, 2B5 is standard conditions and 6w is at a high liquid to gas ratio.

Performance data with high CO_2 inlet concentration for 6 scenarios are specified in Tab. 2 (Sæter, 2021). The original data are taken from Shah *et al*. (2018). These data are from a test campaign with high CO_2 inlet concentration (13.5 vol-%) from a Residue Fluidized Catalytic Cracker (RFCC) at Mongstad. The data are from a campaign called SRD because the purpose was to evaluate the specific reboiler duty under different conditions. The main differences between the scenarios are mainly due to different liquid to gas ratios.

Table 1: Input data for simulations of TCM conditions with low CO_2 inlet concentrations (3-4 vol-%).

Key inputs	Case				
	17F	Goal1	H14	2B5	6w
Lean amin loading (mole CO_2/moleMEA)	0.20	0.20	0.23	0.23	0.25
Lean amin flowrate (kg/hr)	57434	44391	54900	49485	54915
MEA weight% (without CO_2)	31.0	32.3	30.0	31.6	30.4
Flue gas flowrate (kg/hr)	72389	57157	57300	57193	56788
Flue gas pressure (bara)	1.0313	1.0313	1.0313	1.0313	1.0313
Lean amine pressure (bara)	1.0630	1.0630	1.0630	1.0630	1.0630
Flue gas temperature (°C)	29.8	25.0	25.0	28.2	25.0
Lean amin temperature (°C)	37.0	28.6	36.5	36.8	36.9
CO2 removal (%)					
Test result	83.5	90.1	90	87.3	79

Table 2: Key input data and test results for the TCM SRD test cases (high CO_2 inlet concentration).

Key inputs	Case					
	6c	6a	8a	5c	3	4
Lean amin loading (mole CO_2/moleMEA)	0.16	0.19	0.199	0.204	0.251	0.273
Lean amine flowrate (kg/hr)	99670	114873	120360	116455	136867	160821
Flue gas flowrate (Smr/hr)	33908	33900	33934	33918	33699	33874
Flue gas flowrate used (for simplicity) (kg/hr)	43500	43500	43500	43500	43250	43500
Mass ratio lean amine / flue gas	2.29	2.64	2.77	2.68	3.16	3.70
Measured CO_2 removal efficiency (%)	88.3	87.3	87.4	87.3	88.1	85.9

Table 3: Key input data and test results for Campaign 4 test cases with high CO_2 inlet concentration (13.5 vol-%)

Key Inputs	Case			
	1A-1	*1C*	*1D*	*2B*
Lean amin loading (mole CO_2/moleMEA)	0.215	0.29	0.318	0.266
Lean amine flowrate (kg/hr)	120100	200500	200600	165600
Flue gas flowrate used (for simplicity) (kg/hr)	43500	43500	43500	43500
Measured CO_2 removal efficiency (%)	90.1	89.7	78.7	89.4

The temperature was measured at different locations at the same column height for each meter of packing, and the different locations were named A, B, C and D. For the H14 and F17 scenarios, a mean value for each meter of packing was specified in the temperature profile (Hamborg, 2014; Faramarzi 2017).

Data for Campaign 4 from TCM are given in Tab. 3. The original data are from Fosbøl *et al*. (2019). The campaign had the aim of studying lean vapor compression (LVC), but only standard regeneration cases were used as scenarios in this study.

3.2. Specifications for the Equilibrium Based Simulation Tools

The Acid Gas model, which is the recommended equilibrium model in the last Aspen HYSYS version, was used. This is now based on the Electrolyte-NRTL model (Austgen *et al*., 1989), while earlier versions of Aspen HYSYS used other models. The work of Fagerheim (2019) used the

Kent Eisenberg (1976) model. When using Aspen Plus, the Electrolyte-NRTL (Non-Random-Two-Liquid) model was used.

Table 4: Murphree efficiency profiles used in this work.

Stage	Zhu	Zhu_M	Zhu_ Adjusted	0.1 (18 meter)
1	0.23	0.1805	0.310	0.1
2	0.2192	0.1720	0.296	0.1
3	0.2085	0.1636	0.281	0.1
4	0.1977	0.1551	0.267	0.1
5	0.1869	0.1466	0.252	0.1
6	0.18	0.1412	0.243	0.1
7	0.1762	0.1382	0.238	0.1
8	0.1546	0.1213	0.209	0.1
9	0.1438	0.1128	0.194	0.1
10	0.1331	0.1044	0.180	0.1
11	0.1223	0.0960	0.165	0.1
12	0.1115	0.0875	0.150	0.1
13	0.1007	0.0790	0.136	0.1
14	0.09	0.0706	0.121	0.1
15	0.01	0.0078	0.013	0.1
16	0.01	0.0078	0.013	0.1
17	0.01	0.0078	0.013	0.1
18	0.01	0.0078	0.013	0.1
19	0.01			
20	0.01			
21	0.01			
22	0.01			
23	0.01			
24	0.01			

In the Master Thesis work of Zhu (2015), a Murphree efficiency for each of the 24 stages (meter of packing) was estimated for the TCM data set (Hamborg *et al*., 2014). The simplest approach for fitting the E_M profile to the temperature profile was a constant E_M for every stage (Zhu, 2015). Fagerheim (2019) fitted several E_M profiles to the performance data. The Zhu profile has $E_M = 0.01$ on the 10 lowest stages. These profiles were specified in both the Aspen HYSYS and Aspen Plus simulation tools. The different E_M profiles used in this work are presented in Tab. 4.
The Zhu profile is from Zhu (2015). The Zhu_M profile is an adjusted Zhu profile developed by Sæter (2021). The Zhu_Adjusted profile is developed to fit both CO_2 capture efficiency and the temperature profile data from the SRD case 4.

3.3. Specifications for the Rate-Based Tool

The specifications in the rate-based Aspen Plus simulation tool at TCM have been developed for several years, and different versions have been used (Desvignes, 2015; Sætre, 2016). Most of these specifications are based on the work by Zhang *et al*. (2009). Detailed documentation of the rate-based model can be found in the Aspen Plus program documentation. The interfacial area factor was kept constant (as 1.0) in this work. The hold-up factor was varied. The main specifications for the rate based model is given in Tab. 5.

Table 5: Main input to rate-based model (Sæter, 2021).

Common input			
Calculation type	Rate-based		
Fluid properties	RK-ENERTL		
Reaction ID	MEA		
Mass transfer coeff method	BRF-1992 (Bravo-Rocha-Fair)		
Interfacial area method	BRF-1992 (Bravo-Rocha-Fair)		
Holdup method	BRF-1992 (Bravo-Rocha-Fair)		
Number of stages	50		
Film Liquid phase	Discrxn		
No. of discr. points liquid	5		
Film Vapor phase	Film		
Reaction conduction factor	0.9		
Flow model	VPLUG		
Interfacial area factor	1		
Heat transfer coeff method	Chilton and Colburn		
Campaign specific input			
	TCM CHP flue gas	TCM CHP flue gas	Esbjerg cases
Packed height [m]	24	18	17
Packed diameter	3	3	1.1
Packing type	Koch metal 2x	Koch metal 2x	Sulzer Mellapak 2X
Liquid hold up factor	1.0 & tuning	0.72 & tuning	0.72 & tuning
Film discretization ratio	1	2	2

4. Results and discussion

4.1. General Results

The results shown for each model in the scenario figures is the temperature profile. The model parameters (in the case of the rate-based model, the hold-up factor (one parameter)) is adjusted to achieve the specified capture rate. In the case of using an E_M profile, all the E_M values were multiplied with an E_M-factor (one parameter).

4.2. Scenario H14 with equilibrium model

The results from Øi and Fagerheim (2020) are based on the Kent Eisenberg equilibrium model. This work is based on the electrolyte-NRTL model. The temperature profiles for the H14 data in Tab. 2 were calculated in Øi and Fagerheim by fitting the E_M-factor in the simulations to achieve the experimental CO_2 removal. The resulting temperature profiles using the same E_M profiles and E_M-factors are shown in Fig. 2.
The verification shows that the models in this work give approximately the same results as Øi and Fagerheim (2020) using different equilibrium models.

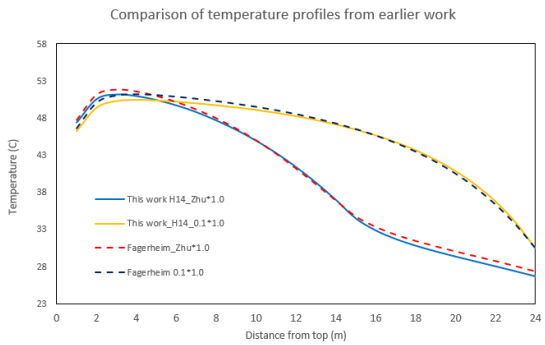

Figure 2: Comparison of measured temperatures for plant data scenario H14 and equilibrium based simulated temperature profiles.

4.3. Scenario H14 with rate-based model

The results in Fig. 3 verifies the simulation of the rate-based model. The interfacial area factor is 1.0, and for this model the hold-up factor is not tuned (equals 1.0). The difference can be explained by Øi and Fagerheim (2020) using the BRF-1985 model and in this work, the BRF-1992 (Bravo *et al*, 1992) is used.

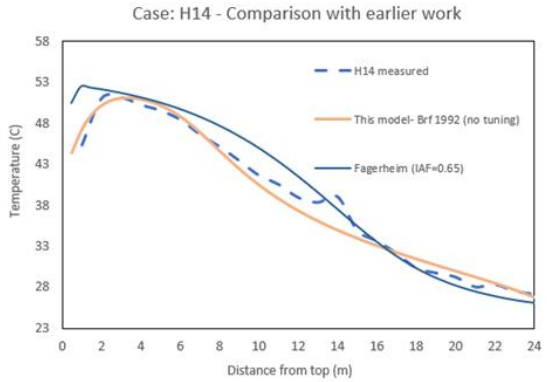

Figure 3: Comparison of measured temperatures for plant data scenario H14 and rate-based simulated temperature profiles.

4.4. Scenario 2B5

The rate-based simulations were fitted to the data from Scenario 2B5. The scenario is for standard conditions at low CO_2 input concentration.

Fig. 4 shows the tuned rate-based model fitted to the CO_2 capture rate for the 2B5 conditions. The interfacial area factor is kept at 1.0, while the hold-up factor is adjusted to 1.6.

When fitting the 5 low CO_2 inlet concentration scenarios, the rate-based adjusted factors (either interfacial area factor or hold-up factor) had to be adjusted more than the E_M-factor for the equilibrium-based models. This was also a general result in Øi and Fagerheim (2020). Especially the H14 and the 6w scenarios are difficult to fit in the rate-based model by adjusting the interfacial area factor.

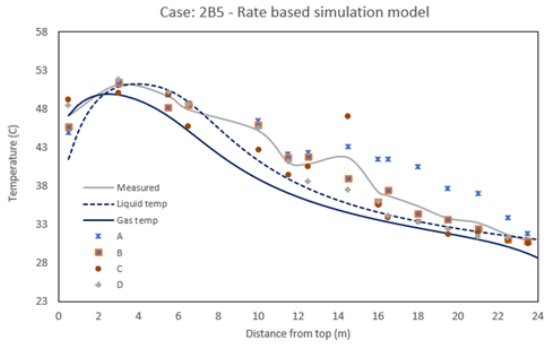

Figure 4: Comparison of measured temperatures for plant data scenario 2B5 and rate-based simulated temperature profiles.

4.5. Scenario SRD Case 4

The 6 scenarios for high CO_2 inlet concentration scenarios specified in Tab. 2 were fitted in the rate-based model by adjusting the hold-up factor and in the equilibrium model by adjusting the E_M-factor. The hold-up factor was adjusted to values between 0.3 and 1.3. The E_M-factor was adjusted to values between 1.33 and 2.05.

All the rate-based scenarios were fitted using 30 wt-% MEA in the input data. Scenario 8a and 3 probably had slightly different MEA concentrations, 28.0 and 30.2 respectively (Sæter, 2021). This resulted in hold-up factors 0.6 and 0.84 with a mean value of 0.72. The value of 0.72 is selected as the hold-up factor for later predictions.

Fig. 5 shows measured and calculated temperatures for Case 4. (CO_2 removal equals test results for all calculations)

The fit is very good when the adjustment factors are used to fit the experimental data. The temperature profiles for rate-based and equilibrium simulations are very similar. In this case, it was tried to adjust the interfacial area factor to fit the CO_2 removal rate, and this was not achievable. Because this was the case also for some of the 5 cases for low CO_2 inlet concentration, adjusting of the hold-up factor was used in this work.

4.6. Scenarios from Campaign 4

The 4 scenarios for high CO_2 inlet concentration specified in Tab. 3 were fitted in the rate-based model by adjusting the hold-up factor and in the equilibrium model by adjusting the E_M-factor. The hold-up factor was adjusted to values between 0.2 and 0.85. The E_M-factor was adjusted to values between 1.72 and 3.4.

For comparison, the highest (Case 1C) and lowest (Case 1A-1) temperature profiles from the rate-based model calculations are included with dotted lines in Fig. 6, together with the results from the HYSYS E_M model. The comparison shows that the two models agree very well for all four cases.

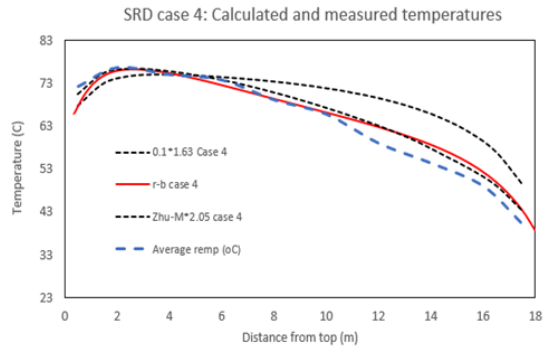

Figure 5: Calculated (equilibrium based and rate-based) and measured temperatures for SRD case 4.

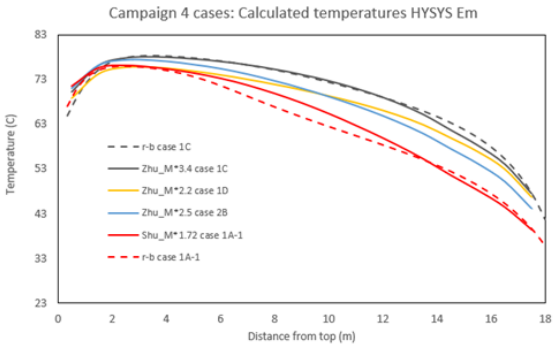

Figure 6: Calculated temperatures for the selected Campaign 4 test cases with the HYSYS E_M model using the Zhu-M profile as a basis. The rate-based model calculations for cases 1C and 1A-1 are included for comparison.

The Zhu E_M profile with an E_M-factor of 1.72 is found to give a good fit to CO_2 removal efficiency as indicated in Fig. 6. The value of 1.72 is selected as the E_M-factor for later predictions in this work.

4.7. Esbjerg cases (4 Cases)

Experimental data were compared to rate-based and equilibrium-based simulations. The purpose of this subsection is to find out whether rate-based and equilibrium-based models fitted to TCM conditions are able to predict CO_2 removal rate and temperatures at Esbjerg conditions.

Table 6: Key input data from Esbjerg test cases and CO_2 removal results.

Key inputs	Case			
	E-1	E-2	E-3	E-4
Lean amin loading (mol CO_2 / mol MEA)	0.29	0.258	0.222	0.181
Lean amine flowrate (m3/hr))	24000	21000	18000	15000
Flue gas flowrate (Nm3/hr)	4952	4975	4999	4999
Mass flow ratio lean amine /flue gas	3.78	3.30	2.83	2.36
CO_2 removal (%)				
Test result	88	90	88	87
Rate-based (lhuf= 0.72)	86.7	88.7	88.8	86.7

For the rate-based model with hold-up factor 0.72, the predicted CO_2 removal rates are given in Tab. 6. The CO_2 removal predictions for all 4 cases are very good.

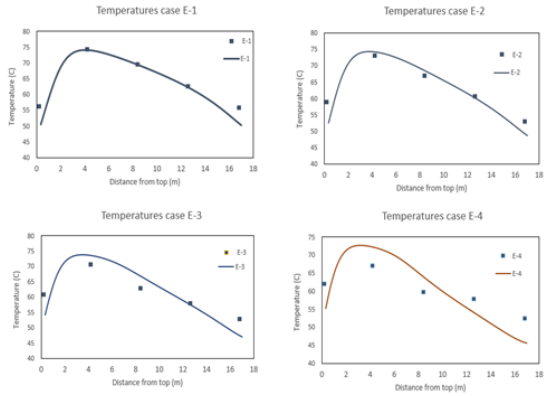

Figure 7: Rate based calculated temperatures compared with measurements for the Esbjerg cases. (Liquid hold-up factor 0.72 for all calculations).

Rate-based simulation results of the temperature profiles are presented in Fig. 7. The predictions of the temperature profiles are very good for case E-1 and E-2 but not so good for E-3 and E-4.

For the equilibrium-based model with the "Adjusted Zhu model", the predicted CO_2 removal rates were 86.1, 89.5, 91.8 and 93.4 and should be compared with the experimental values in Tab. 6. The predictions for E-1 and E-2 are very good. The predictions are not especially good for E-3 and E-4.

Predictions of the temperature profiles for case E-1 and E-2 are given in Fig. 8 and 9. The differences in the top and bottom can be explained by the definitions of simulated temperatures and also the measurement location at top and bottom conditions.

As for the rate-based models, the predictions are very good. The temperature profiles from the equilibrium-based models for E-3 and E-4 (not shown here) are very similar to the rate-based temperature profiles.

For the cases E-3 and E-4, the efficiency was overpredicted (or the E_M values were overpredicted). This overprediction can be explained by stating that Case 3 and 4 were from cases with low amine rate and high rich amine concentrations. At these conditions it is expected that the Murphree efficiencies will be reduced. The rate-based model probably has this efficiency decrease as an integrated part of the model.

4.8. General discussion

The results from this and earlier work show that it is possible to fit both CO_2 removal rate and the temperature profile using either a rate-based or an equilibrium-based model. A model based on other conditions can in most cases be fitted to new conditions by adjusting only one parameter. This

can be the interfacial area factor or the liquid hold-up factor in a rate-based model or an E_M-factor (adjusting all the stage efficiencies in an E_M profile) for an equilibrium-based model.

When trying to use a model fitted at a low CO_2 inlet concentration to predict CO_2 removal rate at a high CO_2 inlet concentration, both the rate-based and the equilibrium-based models had to be heavily adjusted.

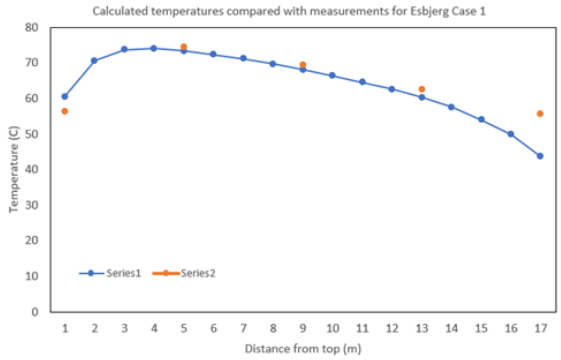

Figure 8: Calculated temperatures compared with measurements for the Esbjerg Case 1 (E_M-factor 1.72).

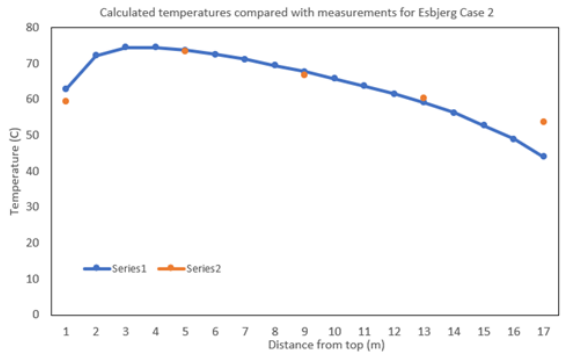

Figure 9. Calculated temperatures compared with measurements for the Esbjerg Case 2 (E_M-factor 1.72).

The rate-based model fitted at TCM conditions for high CO_2 inlet concentration was able to predict the Esbjerg conditions very well. For some cases, also the equilibrium-based model was able to predict the Esbjerg conditions well. When the lean amine flowrate and the lean amine loading changed much, the predictions based on the equilibrium-based model were not good. The dependence of lean amine loading and lean amine flowrate was better predicted by the rate-based model. These dependencies are probably modelled reasonably in the rate-based model and not at all in the equilibrium-based model in this work.

In Øi *et al*. (2018) and in Øi and Fagerheim (2020) it was found that at their conditions (low inlet CO_2 concentration) the equilibrium-based models and rate-based models performed about equally well. This work indicates that for predicting performance at very different conditions, a rate-based model performs better. As stated in Øi and Fagerheim (2020), when the knowledge of the factors used in rate-based simulations becomes better known, the rate-based models can probably be made more predictive.

5. Conclusions

Performance data at TCM have been simulated in both a rate-based model in Aspen Plus (RateSep) and an equilibrium-based model in Aspen HYSYS. The rate-based model's performance data were fitted by only changing the liquid hold-up factor to obtain the experimental CO_2 removal efficiency. In the equilibrium-based model, a Murphree efficiency was specified to fit the CO_2 removal efficiency and the temperature profile. The specified E_M profiles were then used to fit performance data for other scenarios by adjusting only an E_M-factor. The performance (CO_2 removal and temperature profile) was reasonably simulated for each given scenario for all the models. It is shown in this work that the use of the liquid hold-up factor (and not the interfacial area factor) is convenient to fit the rate-based model to performance data. Using fitted parameters at low concentration to predict performance at high inlet CO_2-concentration conditions needed an adjustment factor (liquid hold-up factor or E_M-factor) to obtain correct CO_2 removal predictions. A liquid hold-up factor of 0.72 and an E_M-factor of 1.72 fitted to performance data for high CO_2 concentration at TCM gave reasonable predictions compared to performance data for high CO_2 concentration from the Esbjerg pilot plant.

This work indicate that it is not expected that models fitted to performance data can be used to predict performance at very different conditions. It is however showed that adjusting only one parameter in either a rate-based or an equilibrium based model can give a good fit.

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