Simulation Model for an Amine-based CO_2 Capture Rig

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Abstract: The amine-based CO_2 capture rig at USN in Porsgrunn has been operating since 2007. In this study, the main aim was to develop predictive models in Aspen HYSYS and Aspen Plus for the CO_2 test rig. The models accuracy were verified by comparing different test scenarios with results from the models. Aspen HYSYS and Aspen Plus have simulated eleven scenarios (test series) with varying process parameters. In Aspen HYSYS, Murphree efficiencies (stage efficiences) were fitted, and in Aspen Plus two approaches were used, fitting the interfacial area or the holdup factor to minimize the deviation between the model and experimental data. The Aspen HYSYS model with the fitted Murphree efficiencies (from top to bottom 0.11, 0.1, 0.09 and 0.07) predicted seven scenarios with an average deviation of 12-24 % from experimental data. In the Aspen Plus rate-based model with interfacial area fitted, most of the scenarios were predicted by a model with correlation Brf-85 (mass transfer), Brf-85 (heat transfer) and an interfacial área factor of 0.5. Minimum and maximum deviations for different scenarios were 2.1 and 9 %. In the approach with fitting of the holdup factor, the Brf-92 holdup method with a holdup factor of 0.5 gave the best fit, resulting in an average deviation of 1.4-9 % from the test results across all scenarios.

Keywords: CO_2, amine, rate-based, absorption, simulation

1. INTRODUCTION

The amine-based CO_2 capture rig at the University of South-Eastern Norway (USN) in Porsgrunn has been operating since 2007. It is a small-scale CO_2 capture plant that includes an absorber, a desorber with reboiler and condenser, heat exchangers and pumps. Performance data measuring especially CO_2 capture rate at different conditions using 30 weight-% MEA have been presented in several papers (Øi et al., 2013; Øi et al., 2015; Øi et al., 2017; Øi et al., 2021).

There have been attempts to fit the experimental data from the CO_2 capture rig to simulation models with limited success by fitting data series to a constant Murphree efficiency and using rate-based models. However, the models fitted to experimental data have shown poor predictive properties for conditions outside the experimental range (Karunarathne and Øi, 2019).

Early work on comparing test data with different simulation tools was by Luo et al. (2009) who tested Aspen RadFrac, ProTreat, ProMax, Aspen RateSep, CHEMASIM from BASF and CO2SIM from SINTEF/NTNU using different pilot plant data. The conclusion was that all the models could fit the CO_2 capture rate, but the temperature and concentration profiles were not well predicted.

Much work with fitting CO_2 absorption performance data to Aspen HYSYS or Aspen Plus models have been performed with data from the Test Center Mongstad (TCM). In previous research at USN, data from TCM was used to fit equilibrium models (in Aspen Plus and Aspen HYSYS) to various scenarios by adjusting each stage's Murphree efficiency (E_M), and rate-based models (in Aspen Plus) were fitted by varying the interfacial area factor or the holdup factor (Øi et al., 2018; Øi and Fagerheim, 2020; Øi et al., 2022). Except for this work at TCM, there is very little published information about comparing different models in simulation tools to fit experimental data (Kvamsdal et al., 2011; Razi et al., 2012; Razi et al. 2013a).

In rate-based models, it is traditional to adjust the interfacial area factor to fit the model to performance data, but to use the holdup factor (multiplied with the holdup estimated by in-built correlations) has been used for the same purpose (Øi et al., 2022). Two of the estimation methods in Aspen Plus for several of the correlations, especially the liquid holdup, is Brf-85 and Brf-92 which is short for the Bravo/Rocha/Fair model (Rocha et al., 1992).

This work is based on the Master Thesis of Soudeh Shamshiri (2023) at USN which aims to fit different models in Aspen HYSYS and Aspen Plus to experimental data in the USN test rig. The ultimate goal is to assess which models provide the most accurate predictions for different conditions, and giving the most accurate dependence on the varying process parameters.

This paper starts with an introduction with background literature in section 1. In section 2 different correlations for fitting experimental CO_2 absorption data to models and correlations are presented. In section 3 specifications for the simulations and the data fitting to experimental data from the CO_2 absorption rig are presented. In section 4 the deviations between the simulations and performance data are presented, and the different approaches for modelling and fitting are discussed.

2. METHODS FOR FITTING EXPERIMENTAL CO_2 ABSORPTION DATA TO MODELS

2.1 Equilibrium-based models

Simulation tools include column models for distillation and absorption, which assume equilibrium at each stage. Changing the number of simulation stages until the desired CO_2 capture efficiency is achieved is an easy method of adjusting an absorption equilibrium model to match experimental CO_2 capture efficiency.

Equilibrium based models can be refined by defining a Murphree efficiency at each column stage. This efficiency is calculated as the ratio of the change in mole fraction from a stage to the next divided by the change assuming equilibrium. An absorption column can be defined with a number of stages, e.g. equal to the packed column height, and the Murphree stage efficiency can be specified to be equal for each stage. The constant Murphree efficiency can be fitted to experimental CO_2 efficiency in performance data.

At USN equilibrium models in Aspen Plus and Aspen HYSYS have been fitted to different scenarios (experimental data) at Test Center Mongstad (TCM) by adjusting the Murphree efficiency (E_M) for each stage (Øi et al., 2018; Øi and Fagerheim, 2020; Øi et al., 2022).

2.2 Rate-based models

The process simulation tool Aspen Plus has a rate-based model to describe the mechanisms in the absorption process. Rate-based models calculate rate of mass transfer, rate of heat transfer, pressure drop and equilibrium. Unlike the equilibrium model, the rate-based approach assumes that separation is achieved through mass and energy transfer between gas and liquid which is in equilibrium at the interface. The rate-based modelling is increasingly accepted over traditional equilibrium-stage modelling for CO_2 capture processes.

A rate-based model for CO_2 absorption in MEA is available in the Aspen Plus program. This model was developed and fitted based on data from a CO_2 capture pilot plant at the University of Texas (Zhang et al., 2009). The parameter values in this Aspen Plus model can be changed by the user.

2.3 Correlations in rate-based models

A rate-based model includes correlations (submodels) for mass transfer rates through the gas film and the liquid film, rate of heat transfer, pressure drop, interfacial area and liquid holdup. Aspen Plus has several available correlations for each of these quantities. A review of mass transfer models for both random and packed columns is given by Wang et al. (2005). Reviews of rate-based models for CO_2 capture are presented by Kvamsdal et al. (2011), Razi et al. (2012) and Amirkhosrow et al. (2021). Razi et al. (2013) have evaluated how different correlations in rate-based simulations work in Aspen Plus to predict performance from the Esbjerg pilot plant. The prediction of performance data is very dependent on the correlations used. The models differ in accuracy and correct description of sensitivity of operating parameters like temperature, gas flow and liquid flow.

In the Aspen Plus rate-based model, examples of mass transfer correlations are named Brf-85 (Bravo et al, 1985), Hanley (2012) and Brf-92 (Bravo et al, 1993) and interfacial area methods are named Brf-85, Hanley, Brf-92 and Mod-Tsai. Inaccuracy in one of the correlations can be compensated by adjusting that correlation or other correlations by adjusting model parameters. In Aspen Plus, it is possible to compensate in some of the correlations by adding an adjustment parameter. For the interfacial area, the interfacial area factor is used, and for the liquid holdup, the liquid holdup factor is used.

These correlations normally require physical data (like densities and viscosities) as input. The effect of this is discussed in Nookuea et al. (2015) and Karunarathne and Øi (2019). In this work, the default property methods from the sample file from Aspen Plus are used.

2.4 Equilibrium models

Both an equilibrium-based model and a rate-based model is dependent on the vapour/liquid equilibrium model. In earlier work, different equilibrium models have been used (Øi et al., 2018). Both Aspen Plus and the new Aspen HYSYS acid gas model now use an Electrolyte-NRTL equilibrium model (Austgen et al., 1989). This is used in this work.

3. DATA AND SPECIFICATIONS

3.1. Performance data from test rig at USN

Data were taken from experimental work that has been done for several years in the test rig at USN, a picture is shown in Fig. 1 and a simplified P&ID is shown in Fig. 2. Eleven scenarios have been defined in this work in which different process parameters like gas and liquid flow rate, solvent inlet temperature and CO_2 inlet concentration were varying. Figures 3, 4 and 6 are from Øi et al. (2017), Figure 5 is from Øi et al. (2013) and Figure 7 is from Øi et al. (2021).

Fig. 1. Picture of CO_2 test rig at USN.

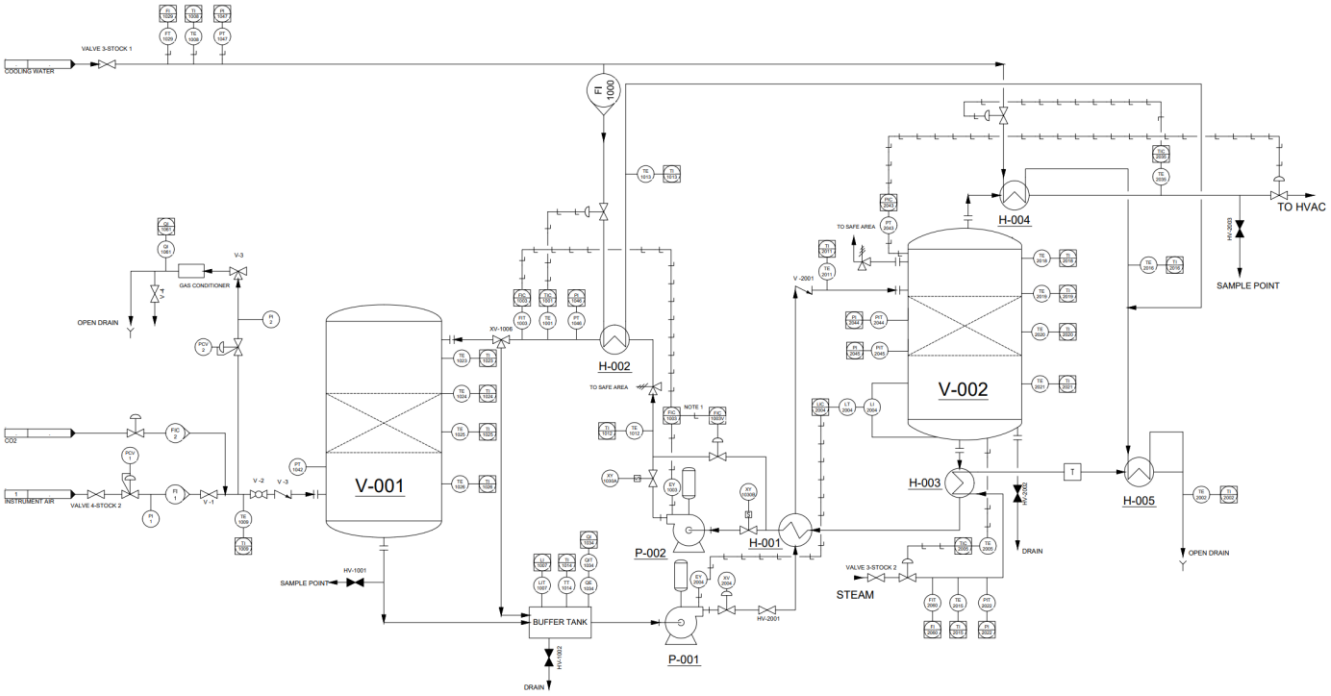

Fig. 2. Simplified P&ID for USN test rig.

Figures 3 to 7 show data as a function of only one parameter. The CO_2 capture rate is dependent on many parameters. Detailed information can be found and the exact numerical values for the data points can be found in the original references or in Shamshiri (2023).

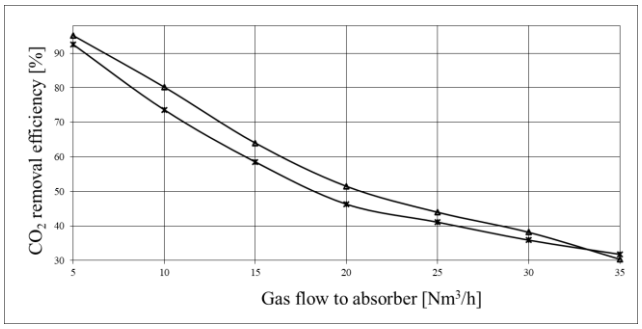

Fig. 3. Data for varying the gas flow at a high liquid flow in scenario 1-1 and 1-2. The upper curve is for liquid flow 150 l/h and the lower curve is for 140 l/h. There is 10 % CO_2 in the inlet gas.

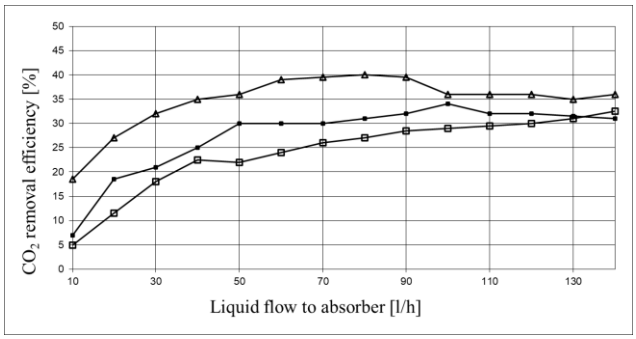

Fig. 4. Data for varying liquid flow in scenario 2-1, 2-2 and 2-3. The upper curve is for 5 % CO_2 in inlet gas, the middle curve for 10 % CO_2 and the lower curve is for 15 % CO_2.

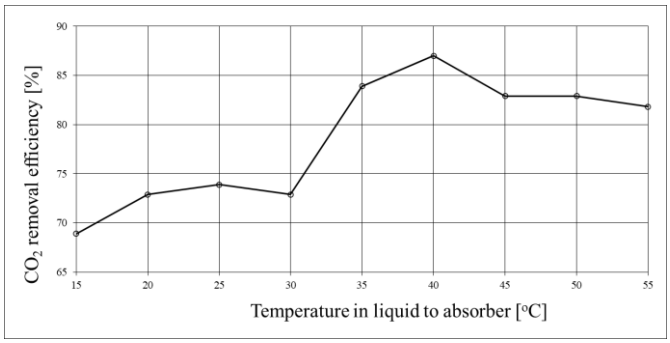

Fig. 5. Data for varying the temperature in scenario 3-1. The data are for constant gas flow 14 Nm^3/h and liquid flow 50 kg/h. There is 10 % CO_2 in the inlet gas.

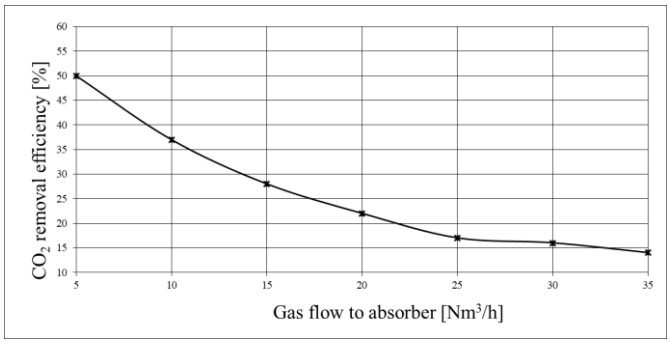

Fig. 6. Data for varying the gas flow at a low liquid flow for scenario 4-1. Liquid flow is 20 l/h and there is 10 % CO_2 in inlet gas.

The selection of scenarios is tried to be representative for the available data for the different variables and for the variable ranges. The selection have not been systematical, and in this work there has not been any evaluation about which experimental data for the test rig which are of highest quality.

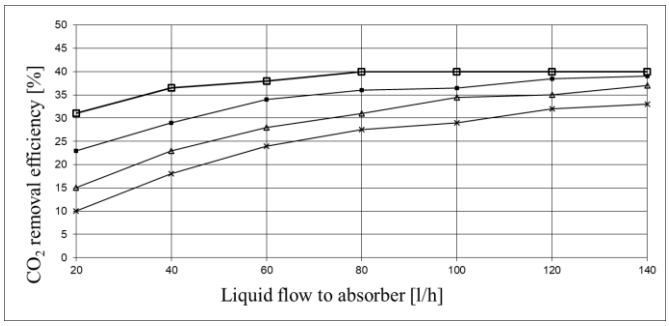

Fig. 7. Data for varying the liquid flow for scenario 5-1, 5-2, 5-3 and 5-4. The curves are from top to bottom for 2.5 % CO_2 in inlet gas, 5 %, 10 % and 15 %. Gas Flow is 30 Nm^3/h.

3.2. Specifications for the equilibrium-based simulation tools

The input specifications in Table 1 for the Aspen HYSYS absorber model are similar to earlier simulations (Øi et al., 2018; Øi and Fagerheim, 2020). A flowsheet of the process is shown in Fig. 8.

Table 1. Specifications for equilibrium-based calculations.

Specifications – Aspen HYSYS equilibrium	
Calculation Type	equilibrium
Property method	Acid gas- Chemical solvents
Valid phases	Vapor-Liquid
Number of stages	4
Nominal pressure	110 kPa
Uniform section	yes
Internal type	Packed
Solving method	Modified HYSIM Inside-out

3.3. Specifications for the rate-based tool

The input specifications for the rate-based absorber calculations are given in Table 2. A flowsheet is shown in Fig. 9. In the rate-based simulation in Aspen Plus, the local model example "ENRTL-RK_Rate_Based_MEA_Model" from the Aspen library was used. The Elec-NRTL thermodynamic package with Redlich-Kwong equation of state (RK) was chosen.

Table 2. Specifications of the model for rate-based calculations

Specifications – Aspen Plus rate-based	
Calculation Type	Rate-based
Property method	ENRTL-RK
Henry comp ID	Global
Chemistry ID	MEA-CHEM
Valid phases	Vapor-Liquid
Number of stages	4
Hold up factor	0.5-1
Reaction condition factor	0.9
Packing type	Mellapak 250Y
Section diameter (m)	0.1
Section packed height (m)	1.5
Flow model	VPlug*
Interfacial area factor	0.5-1-1.3-1.6
Film liquid phase	Discretize film
Film vapor phase	Consider film
Mass transfer coefficient method	Brf-85/Hanley/Brf-92
Heat transfer coefficient method	Chilton and Colburn
Interfacial area method	Brf-85/Hanley/Brf-92/Mod-Tsai
Holdup method	Brf-92
Add. Discretize points liquid	5

The specifications for the Aspen Plus model are mostly as in earlier work (Øi et al., 2021), but the chosen correlations, and especially the combination of correlations, are mostly a result of trial and error. A combination of correlations obtaining good prediction over a large range of the varied parameter is regarded as a useful method. The choice of interfacial area is traditional (Zhang et al., 2009; Øi et al., 2018; Øi and Fagerheim, 2020), but the use of the holdup factor as fitting variable is only found in earlier work by Øi et al. (2022).

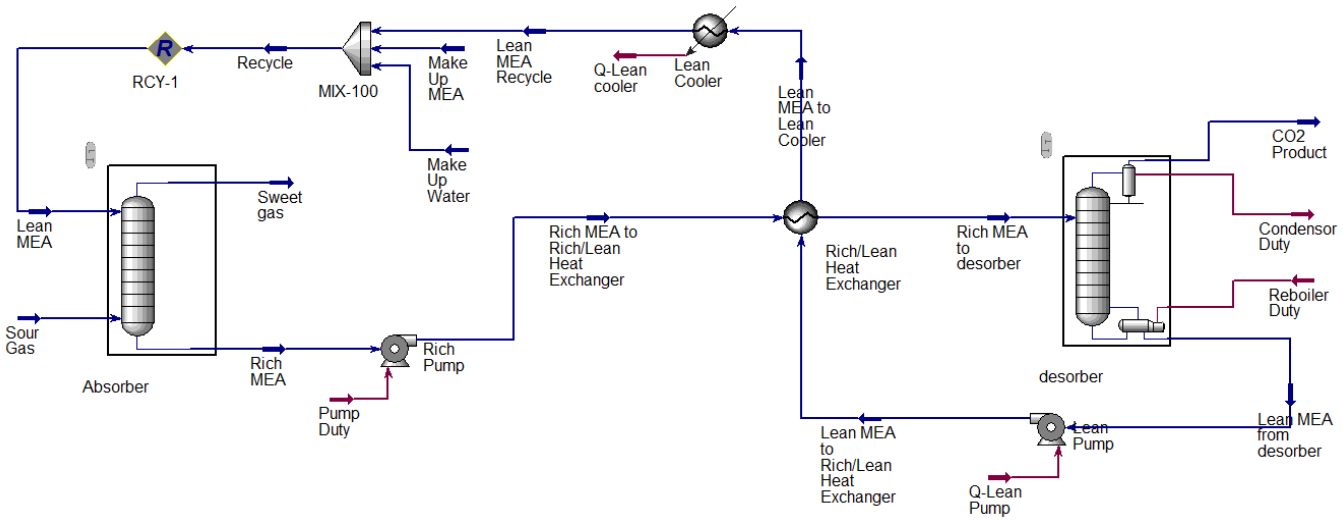

Fig. 8. Process simulation model of the CO_2 capture rig in Aspen HYSYS.

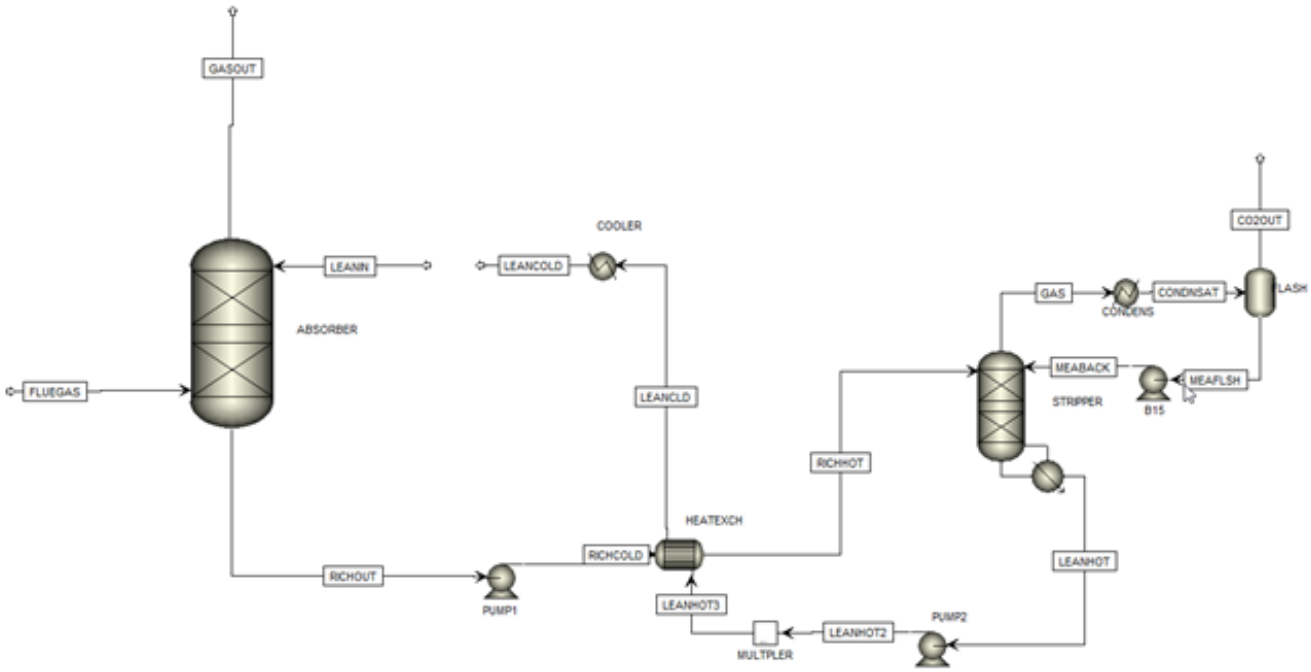

Fig. 9. Process simulation model of the CO_2 capture rig in Aspen Plus.

3.4. Simulations to fit experimental values

The rate-based model developed in the simulation focused on the absorber column. In Aspen HYSYS, the Murphree efficiency was adjusted to determine the experimental CO_2 removal efficiency for a given scenario. Using Aspen Plus, the interfacial area factor or the holdup factor were fitted to obtain the experimental CO_2 removal efficiency.

3.5. Simulations to predict performance

Based on experimental data, the fitted models in Aspen HYSYS or Aspen Plus were used to predict conditions at different operating conditions The deviation between predicted and measured CO_2 capture rate was calculated in %.

4. SIMULATION RESULTS AND DISCUSSION

4.1. General Results

One fitted simulation case is the result for each data point in each of the eleven scenarios. The main result from each simulation is the CO_2 removal rate which is found as the CO_2 amount in the gas feed to the absorber minus the CO_2 amount in the gas from the absorber. The deviation in this work is the simulated CO_2 removal minus the experimental CO_2 removal divided by the simulated value.

4.2. Simulation model for equilibrium-based model

The equilibrium-based model with defined parameters and properties has been simulated for the eleven scenarios in the test rig USN. The results in Fig. 10 show that the Aspen HYSYS can predict the CO_2 removal rate for the sets of scenarios 2-1, 2-2, 2-3, 5-1, 5-2, 5-3 and 5-4 where the deviation of predicted model from experimental data is between 12-24%. In these simulations the Murphree efficiency for all stages were defined to be 0.11, 0.1, 0.09 and 0.07 from top to bottom.

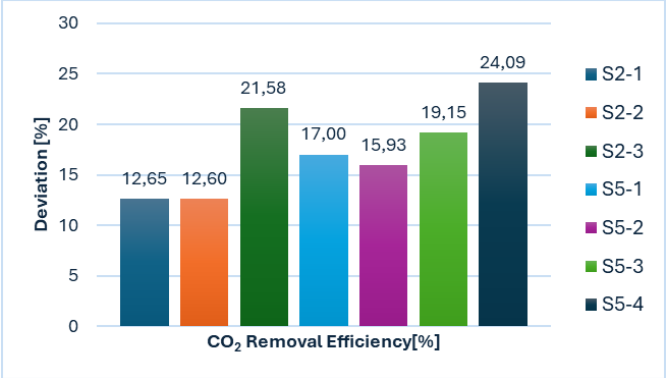

Fig. 10. Deviation between the experimental data and the simulation results from Aspen HYSYS.

4.3. Simulation model for rate-based model

The model was used to simulate the performance data using various mass transfer and interfacial área correlations. Figure 11 shows that the Brf-85 method for both mass and heat transfer and an interfacial area factor of 0.5 or 1.0 gives smaller deviation than when the Hanley method is used. When the interfacial area factor is changed from 1 to 0.5, most of the experimental data will be reasonably predicted when varying the process parameters.

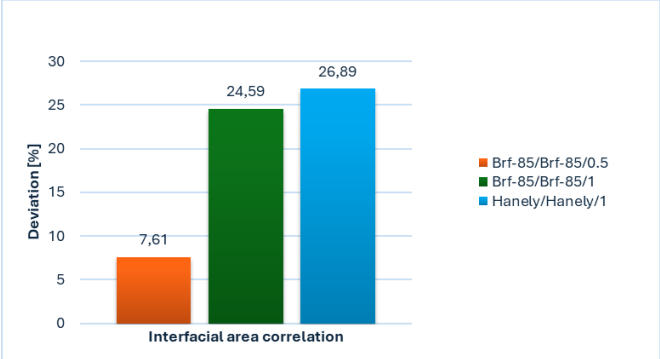

Fig. 11. Deviation of simulated data in Aspen Plus/Interfacial area from experimental data with different correlations for scenario 2-1.

4.4 Deviation results for rate-based model

If the Brf-85 correlation could not predict all experimental data, another method would have simulated all scenarios, such as changing the liquid holdup factor. According to Fig. 12, Brf-85 as a mass transfer and interfacial area method with factor 1 beside Brf-92 as a holdup correlation with holdup factor 0.5 provides results with less error tan the Hanley method and the same holdup correlation and factor.

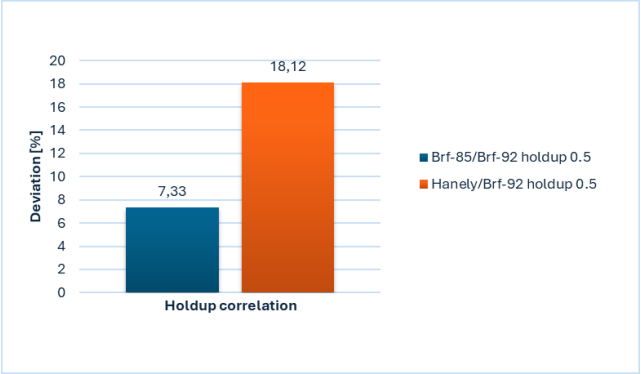

Fig. 12. Deviation of simulated data in Aspen Plus/Holdup from experimental data with different correlations for scenario 2-1.

Figures 11 and 12 indicate that using the interfacial area or holdup factor as the variable parameter gives similar sum of deviations at least for this case.

Only a limited number of different correlations and especially combination of correlations were tested. Evaluation of other correlations is discussed in the general discussion.

4.5 Deviation results for rate-based model with varying interfacial area

In Fig. 13, the deviations are shown for all scenarios when using a model with Brf-85 for both the heat transfer, mass transfer and the interfacial área and an interfacial area factor 0.5, except for scenarios S3-1 and S4-1. In S3-1 the Hanley model for interfacial área and an interfacial area factor of 1.3 was necessary to fit the data and in S4-1 the Brf-92 model for heat and mass transfer, the ModTsai model for interfacial area and an interfacial área factor of 1.0 were necessary to fit the data satisfactory.

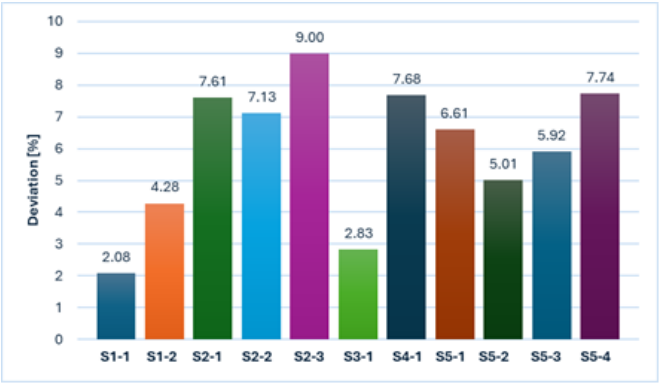

Fig. 13. Deviation between the experimental data and the simulation results from Aspen Plus/Interfacial area method with different correlations for S3-1 and S4-1.

For the scenarios S3-1 and S4-1, using the model with Brf-85 and interfacial area = 0.5 the fit was not satisfactory. Øi and Fagerheim (2020) and Øi et al. (2022) experienced that when fitting TCM data to a similar model, some experimental data was difficult to fit using the Brf-85 model for interfacial area, so the Brf-92 was used by Øi et al (2022).

4.6 Deviation results for rate-based model with varying holdup factor

Figure 14 shows the deviation results for all the eleven data sets or scenarios. The same combination of correlations models were used in these simulations, with Brf-85 for heat and mass transfer, Brf-92 for holdup and h = 0.5.

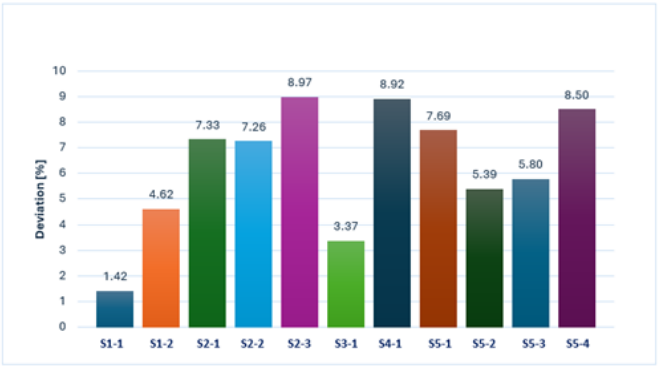

Fig. 14. Deviation between the experimental data and the simulation results from Aspen Plus/Holdup method with the same correlations also for S3-1 and S4-1.

By running the simulation for all the experiments, the average deviation from all sets of scenarios is below 9 %.

4.7 General discussion

The results from this and earlier work show that it is possible to fit the CO_2 removal rate for a performance data set using either a rate-based or an equilibrium-based model. For most cases, it is enough to adjust only one parameter. This can be the interfacial area factor or the liquid holdup 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.

In Luo et al. (2009), Øi et al. (2012) and in Øi and Fagerheim (2020) it was claimed that at their conditions (low inlet CO_2 concentration) the equilibrium-based models and rate-based models performed about equally well in fitting the available performance data. This work indicates that for predicting performance at very different conditions, a rate-based model performs better than an equilibrium-based model.

This work and the work by Øi et al. (2022) show that a promising approach to obtain a predictive model for a CO_2 capture plant based on performance data, is to vary the holdup factor to fit performance data. It is recommended for future work to compare the approach of fitting the interfacial area factor and the holdup factor in more detail.

There is little in the literature about which combination of correlations for mass transfer, heat transfer, interfacial area and holdup that gives the most accurate prediction as a function of varied temperature, gas flow and liquid flow. This topic is discussed by Kvamsdal et al. (2011), Kvamsdal and Hillestad (2012), Razi et al. (2012) and Reza et al. (2013b). To find the most accurate or most convenient combination of correlations for rate-based simulation of amine-based CO_2 capture is a challenge for future work. One strategy is to test all (or most of) the possible combinations of models. Then it is a challenge to treat large amounts of experience data to obtain an optimum choice of models. Another strategy is to study the correlations more in detail to select the most accurate correlations. Then it is a challenge whether to choose the most accurate correlations, or the correlations which give the most accurate dependence on the varying process parameters.

5. CONCLUSIONS

In this study, the main aim was to develop predictive models in Aspen HYSYS and Aspen Plus for the CO_2 test rig. Aspen HYSYS and Aspen Plus have been used to simulate eleven scenarios (test series) with varying process parameters. In Aspen Plus two approaches were used, fitting either the interfacial área factor or the holdup factor to minimize the deviation between model and experimental values. An Aspen HYSYS model could not predict all experimental data when varying liquid temperature, inlet gas temperature and flow rates. A fitted rate-based model in Aspen Plus gave less error than the equilibrium-based Aspen HYSYS model. In the rate-based model with interfacial area fitted, most scenarios can be predicted by a model with correlation Brf-85 (mass transfer), Brf-85 (heat transfer) and an interfacial factor of 0.5. Minimum and maximum deviations for different scenarios were 2.1 and 9 %. In the approach with fitting the holdup factor, the Brf-92 holdup method with a holdup factor of 0.5 gave the best fit, resulting in an average deviation of 1.4-9 % from the test results for all scenarios. Testing and evaluation of combinations of correlations for rate-based models is a future challenge.

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