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Improving Prediction Accuracy of a Rate-Based Model of an MEA-Based Carbon Capture Process for Large-Scale Commercial Deployment

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ABSTRACT

Carbon capture and storage (CCS) technology will play a critical role in reducing anthropogenic carbon dioxide (CO_2) emission from fossil-fired power plants and other energy-intensive processes. However, the increment of energy cost caused by equipping a carbon capture process is the main barrier to its commercial deployment. To reduce the capital and operating costs of carbon capture, great efforts have been made to achieve optimal design and operation through process modeling, simulation, and optimization. Accurate models form an essential foundation for this purpose. This paper presents a study on developing a more accurate rate-based model in Aspen Plus® for the monoethanolamine (MEA)-based carbon capture process by multistage model validations. The modeling framework for this process was established first. The steady-state process model was then developed and validated at three stages, which included a thermodynamic model, physical properties calculations, and a process model at the pilot plant scale, covering a wide range of pressures, temperatures, and CO_2 loadings. The calculation correlations of liquid density and interfacial area were updated by coding Fortran subroutines in Aspen Plus®. The validation results show that the correlation combination for the thermodynamic model used in this study has higher accuracy than those of three other key publications and the model prediction of the process model has a good agreement with the pilot plant experimental data. A case study was carried out for carbon capture from a 250 MW_e combined cycle gas turbine (CCGT) power plant. Shorter packing height and lower specific duty were achieved using this accurate model.

1. Introduction

1.1. Background

Increasing concentrations of greenhouse gases (GHGs) caused by anthropogenic activities are responsible for most of global warming [1]. Carbon dioxide (CO_2) is a main GHG, accounting for 76% of total GHG emissions in 2010 [2]. The International Energy Agency (IEA) set up a BLUE Map scenario with 14 Gt of CO_2 emissions in 2050 compared with 57 Gt of CO_2 emissions in the baseline scenario [3]. In order to achieve this target, carbon capture and storage (CCS) technology will play a vital role in delivering 19% of cumulative CO_2 emission reductions between 2015 and 2050 in the power sector [3].

Among the three main approaches envisaged for CO_2 capture from power plants—pre-combustion capture, post-combustion capture, and oxyfuel capture [4]—the solvent-based post-combustion carbon capture (PCC) process is regarded as the most promising technology for commercial deployment [5,6]. In solvent-based carbon capture technology, CO_2 is separated from flue gas after combustion by chemical absorption; monoethanolamine (MEA) is regarded as a benchmark solvent for this process.

1.2. Previous studies

A complex electrolyte aqueous solvent is involved in the MEA-

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based PCC process [7], which requires accurate thermodynamic modeling and physical properties calculations for its modeling. Thermodynamic data, especially regarding CO_2 solubility, have been reported for 30 wt% MEA aqueous solutions [8,9] and for a wider MEA solution concentration range [10,11]. For the parameterization and validation of physical properties calculation methods of an MEA-H_2O-CO_2 mixture, experimental data on MEA aqueous solutions are valuable, especially with different CO_2 loading. Correlations for the calculation of the density and viscosity of MEA-H_2O-CO_2 mixtures at different temperatures and MEA concentrations can be found in the literature [12–14]. In terms of mass transfer and thermal performance of the integrated MEA-based PCC process, several experimental campaigns [15,16] have been conducted.

For a highly nonlinear electrolyte MEA-H_2O-CO_2 solution, the electrolyte non-random two-liquid (eNRTL) model [17,18] is the most widely adopted model [10,19]. Recently, some studies [20,21] have also used the perturbed-chain statistical association fluid theory (PC-SAFT) [22,23] equation of state (EOS) for the vapor phase of an MEA-H_2O-CO_2 mixture, with a system temperature of up to 500 K and a system pressure of up to 15 MPa.

For this capture process, significant energy is consumed for solvent regeneration [6]. Thus, the cost of carbon capture is high when PCC is added to the emitters. Great research efforts have been taken to reduce the carbon capture cost through process modeling and simulation approaches. Most early studies were carried out for the parametric sensitivity analysis of solvent-based PCC processes in the context of coal-fired power plants [24–27]. Some studies were carried out on integrations between power plants and carbon capture plants [28–30]. Several studies focused on optimizing the whole plant through process optimization [31–34].

However, obvious inconsistencies in the literature were found for key equipment design features and key operational variables. For example, the packing height varies from 13.6 m [35] to 30.6 m [32] for the absorber and from 7.6 m [35] to 28.15 m [21] for the stripper for similar capture tasks. The optimal lean loading range is equally wide from 0.132 $mol_{CO_2}\cdot mol^{-1}_{MEA}$ [31] to 0.234 $mol_{CO_2}\cdot mol^{-1}_{MEA}$ [36], with corresponding specific duty in a range from 3.77 $GJ\cdot t^{-1}_{CO_2}$ to 4.35 $GJ\cdot t^{-1}_{CO_2}$. Those inconsistencies cause confusion for future research in this field. They may also cause some trouble for the engineering design of a large-scale commercial deployment.

The main reasons for the abovementioned knowledge gaps may be related to conflicts between the complexity of the integrated system and the accuracy requirement of the modeling and simulation studies. Firstly, the models used in some publications were relatively simple. For example, equilibrium models were used for the mass transfer and reaction in both the absorber and the stripper [37]. For a rate-based model, the correlations for calculations of mass transfer coefficients, interfacial area, liquid holdup, and pressure drop inside packing beds also have a large impact on the prediction accuracy [38,39]. For the kinetics-controlled reactions, it is found that the values of the kinetics of reverse reactions for bicarbonate formation are different for the absorber and the stripper [40]. Inappropriate correlations used in the models would significantly affect the accuracy of model predictions.

1.3. Aim and novel contribution

In order to address the abovementioned knowledge gaps, this study aims to improve the accuracy of the rate-based model in Aspen Plus® for the MEA-based carbon capture process. The novel contributions of this paper can be justified by the following: ① A new combination of correlations was selected after comparing model predictions with the experimental vapor-liquid phase equilibrium (VLE) data; ② the correlations for predicting the liquid density of the mixture and the effective vapor-liquid interfacial area were improved by coding Fortan subroutines in Aspen Plus®; ③ different kinetics parameters were used for reverse reactions for bicarbonate formation in the absorber and the stripper, respectively, thus reflecting the nature of the different operating conditions in the absorber and the stripper; and ④ the rate-based process model was validated with the experimental data and pilot plant data at three different stages, including thermodynamic modeling, physical properties calculations, and process model development at the pilot scale.

2. Framework of modeling of the solvent-based carbon capture process

Using an amine solvent to absorb CO_2 from exhaust gases is a reactive absorption process involving an electrolyte aqueous solvent [6]. The modeling of this non-ideal multi-component system is a systematic work at different levels. Fig. 1 outlines the modeling framework for such a PCC process. Although the software package Aspen Plus® was used for the modeling and simulation of the process, it is important to check the calculation methods with their corrections in order to ensure the accuracy of the process simulation and optimization.

Accurate prediction of the physical properties of pure components and mixtures is one of the basic prerequisites in process modeling and simulation. As the first step, the thermodynamic model should be developed to present VLE and to calculate the state parameters of the MEA-H_2O-CO_2 mixture, such as the temperature, pressure, and composition of the liquid and vapor phases. The solubility of CO_2 in the MEA-H_2O-CO_2 mixture is a key parameter, and is normally used for validation purposes for the calibration of the correlations or for selection for VLE calculation.

The physical properties are part of the correlations for heat transfer, mass transfer, interfacial area, liquid holdup, and pressure drop. It is important to choose the right physical property models to ensure the success of process modeling and simulation.

At the process level, both absorption and desorption in the packed columns are key processes. A rate-based model offers better accuracy than an equilibrium model for the absorption performance of the columns [41]. This accuracy is a function of the appropriate correlations used for liquid and vapor phase mass transfer coefficients, the effective vapor-liquid interfacial area, and the pressure drop in the rate-based model.

This framework shows that the rate-based model for this solvent-based carbon capture process is a highly nonlinear model, which has numerous parameters, correlations, and equations. Therefore, it is not realistic to completely repeat the published models with the same input conditions. This is also the main consideration behind the choice to use three-stage validations in this study, and to update some correlations by coding a Fortran subroutine in Aspen Plus® to ensure model accuracy, rather than directly comparing the process performance with those of other published models. Using this three-stage model validation method, the model was dissected in detail based on the logical structure of numerical modeling, allowing more insights to be obtained.

3. Thermodynamic modeling of the MEA-H_2O-CO_2 system

3.1. EOSs and relevant model parameters

In this study, the PC-SAFT EOS [22,23] is used to calculate the properties of the vapor phase, and the eNRTL [18] method is used to model the electrolyte system of an MEA-H_2O-CO_2 mixture.

3.1.1. The PC-SAFT EOS for the vapor phase

Compared with some typical cubic EOSs such as the Peng-Robinson