International Journal of Greenhouse Gas Control 34 (2015) 117–128

Contents lists available at ScienceDirect

International Journal of Greenhouse Gas Control

journal homepage: www.elsevier.com/locate/ijggc

Aspen Plus rate-based modeling for reconciling laboratory scale and pilot scale CO_2 absorption using aqueous ammonia

Jialin Liu [a], Han-Ci Gao [b], Chao-Chin Peng [b], David Shan-Hill Wong [b,*], Shi-Shang Jang [b], Jui-Fu Shen [c]

[a] Department of Chemical and Materials Engineering, Tunghai University, No. 1727, Sec.4, Taiwan Boulevard, Taichung, Taiwan, ROC
[b] Department of Chemical Engineering, National Tsing Hua University, No. 101, Section 2, Kuang-Fu Road, Hsinchu, Taiwan, ROC
[c] New Materials Research & Development Department, China Steel Corporation, 1, Chung Kang Road, Hsiao Kang, Kaohsiung, Taiwan, ROC

ARTICLE INFO

Article history:
Received 8 August 2014
Received in revised form
13 December 2014
Accepted 19 January 2015
Available online 7 February 2015

Keywords:
Post combustion capture
Aqueous ammonia
Process simulation
Rate-based model

ABSTRACT

In this study, simulations of CO_2 absorption by aqueous ammonia using the rate-based model of Aspen Plus were investigated using laboratory scale experiments and pilot data in the literature. It was found that predictions are greatly influenced by the choice of transfer condition factor and reaction condition factor, mass transfer coefficients, and kinetic parameters in the rate-based model. Using the kinetic data provided by Pinsent et al. (1956), and mass transfer correlations provided by Billet and Schultes (1993), the optimal settings of transfer and reactor condition factors were found to be 0.75 and 0.25, respectively, by fitting experimental data obtained in the laboratory using 3 wt% aqueous ammonia. This set of parameters can be extended to the laboratory results of using 7 wt% aqueous ammonia, as well as pilot plant data provided by Yu et al. (2011). The results show that it is very important to reconcile the simulation model with data from various operating conditions and different scales before it is used for reliable process design and optimization.

1. Introduction

Post combustion capture (PCC) with chemical absorption is considered to be one of the more suitable techniques to capture CO_2 from flue gases generated by energy intensive process, because PCC can be easily retrofitted to the existing plants. Amine and ammonia-based solvents are two major types of absorbents that have been widely investigated in the PCC research.

Bai and Yeh (1997) reported that an overall CO_2 removal efficiency of over 95% can be achieved, and that the absorption capacity of NH_3 is ca. 0.9 kg-CO_2/kg-NH_3 under appropriate operating conditions. Compared with the absorption performance achieved using monoethanolamine (MEA) absorbents, whose maximum CO_2 removal efficiency and absorption capacity are 94% and 0.4 kg-CO_2/kg-MEA (Yeh and Bai, 1999), respectively, the CO_2 capture ability using NH_3 has been established.

In order to investigate the feasibility of using NH_3 to capture CO_2, several pilot plants have been constructed. For example, the chilled ammonia process (CAP) was developed by Alstom (Gal, 2008). Telikapalli et al. (2011) reported that CAP pilot plants have been installed at the AEP Mountaineer Power Plant in USA and the Test Centre Mongstad in Norway. The former was designed to capture CO_2 at a rate of 270 ton/day from a coal-fired power plant, and the latter was installed to capture 220 ton of CO_2 per day from flue gases generated by a fluid catalytic cracking unit (FCCU) and a gas turbine.

Powspan extended the patented electro-catalytic oxidation (ECO) technology (Duncan et al., 2005), which simultaneously removes mercury, nitrogen oxides (NO_x), and sulfur dioxide (SO_2) from the flue gases of coal-fired power plants, to install an ECO_2 pilot plant at FirstEnergy's R.E. Burger Plant, where the CO_2 capture rate was designed at 20 ton/day and the overall CO_2 removal efficiency was over 90%. In addition, the absorber temperature was approximately 54 °C, thereby eliminating the need for a chill utility for the CO_2 absorber. In order to investigate the feasibility of the ECO_2 process, McLarnon and Duncan (2009) reported that the power penalty of the ECO_2 process is 16% and the energy consumption is 1.1 GJ/ton-CO_2 on the basis of laboratory results, which is 27% of the MEA energy requirement.

CSIRO Australia and Delta Electricity jointly constructed a pilot plant at Munmorah power station (Yu et al., 2011) to evaluate the technical feasibility of PCC using ammonia as an absorbent under real flue gas conditions. They designed seven experimental campaigns in which the absorber temperature was maintained within 15–30 °C, the ammonia concentration was below 6 wt%, and the CO_2 loading of lean solvent was between 0.2 and 0.4.

* Corresponding author. Tel.: +886-3-5735294. fax: +886-3-5725924.
E-mail address: dshwong@che.nthu.edu.tw (D.S.-H. Wong).

http://dx.doi.org/10.1016/j.ijggc.2015.01.009
1750-5836/

The experimental results showed that a CO_2 removal efficiency of over 85% could be achieved even when the concentration of the NH_3 absorbent was under 6 wt%; however, the lowest regeneration energy from the trials was 4–4.2 GJ/ton-CO_2, which is close to the energy consumption required for using 30 wt% MEA. The use of dilute ammonia solutions means that the absorbent has a low CO_2 content, and the stripper pressure was set at 300–850 kPa in the pilot tests; therefore, more than 50% of the regeneration energy was used to heat the solvent to maintain the stripper pressure. In order to control the ammonia slip from the top of the stripper, the condenser was operated at 20–25 °C.

RIST and POSCO jointly performed PCC pilot tests to capture CO_2 from combustion product of blast furnace gas (BFG). Kim et al. (2009) demonstrated that a CO_2 removal efficiency of over 90% could be achieved with the use of as low as 2 wt% NH_3, once a proper packing structure for maximizing the contact area between the gas and liquid was selected. However, in order to minimize the regeneration duties for the CO_2 stripper and the NH_3 concentrator, 5 wt% NH_3 solution was recommended by Kim et al. (2009). Rhee et al. (2011) utilized two water wash sections that were respectively added on the tops of the absorber and stripper in order to control the ammonia slips. They also used a side stream cooler to reduce the absorber temperature to ca. 5 °C and found that the ammonia slip could be reduced from 1000 to 100 ppm while maintaining the CO_2 removal efficiency at over 90%. The pilot trials (Rhee et al., 2011) showed that dilute ammonia solutions can effectively capture the CO_2 emitted from the BFG, and the ammonia slips can be properly controlled. More recently, Han et al. (2014) reported that the minimum overall energy consumption was approximately 4 GJ/ton-CO_2, depending on the operating conditions and the packing types of the CO_2 stripper, from the pilot trial results at the POSCO-Pohang plant. In their report, the energy consumption attributed to the CO_2 stripper can be as low as 2.5 GJ/ton-CO_2. This value is much lower than that of Yu et al. (2011) reported. However, a value of 1.8 GJ/ton-CO_2 was attributed to the concentrator for wash water used for ammonia abatement, and this energy consumptions cannot be entirely decoupled from regeneration, since ammonia produced by the stripper is recycled into the CO_2 stripper.

Although these pilot trials have provided demonstrations for CO_2 capture using NH_3, there remained two critical issues that needs to be resolved: (1) how to control the ammonia slip and (2) the energy expenditure required. A proper simulation model is required for assessing the flowsheet alternatives and optimal operating conditions. However, there are a number of unresolved issues that required further clarifications in the model development. Firstly, it was generally believed that CO_2 absorption in aqueous ammonia is a relatively slow process and is limited by mass transfer in the liquid phase. Puxty et al. (2010) used a wetted-wall column to measure the CO_2 absorption rates using 1–10 wt% ammonia solutions. They found that the overall mass transfer coefficient for aqueous ammonia at 5–20 °C was at least 1.5–2 times smaller than that of MEA at 30–40 °C. However, Darde et al. (2011) measured the absorption rate of CO_2 using 10 wt% NH_3 solvent at 31 °C by applying a wetted-wall column and reported that the absorption rate was comparable to that achieved using 30 wt% MEA at 41 °C. Liu et al. (2011) also utilized a wetted-wall column to measure the reaction rates of CO_2 in 1–7.5 wt% aqueous ammonia at 10–40 °C. They reported that the reaction rate constant of aqueous ammonia was much lower than that of MEA. Despite the general uses of wetted-wall columns for measuring the reaction rates of CO_2 with NH_3, the conclusions were not consistent in the above-mentioned papers. In addition, the reaction rates of CO_2 with aqueous ammonia reported in the literature have been measured using a variety of experimental apparatus. For example, Pinsent et al. (1956) used a mixing chamber and an observation tube, where the temperature of the flowing solution along the tube was measured. The reaction rate constants were decided based on the temperature profile. Derks and Versteeg (2009) utilized an isothermal stirred-cell type reactor to prepare the aqueous ammonia. A predetermined amount of CO_2 was introduced into the reactor from the gas supply vessel, and the CO_2 partial pressure was measured in the reactor to determine the reaction rates. Qin et al. (2010) used a string of disc contactors, where the absorbent was fed into the top of the contactor. The gas mixture was circulated in the direction counter to the liquid flow. A summary of the apparent reaction rates for CO_2 absorption with aqueous ammonia reported in the literature was presented by Jilvero et al. (2014), indicating that the discrepancy between the measured reaction rates is over an order of magnitude. The inconsistency of the reaction rates increases the complexity of developing an absorption model to describe the behavior of PCC using aqueous ammonia. However, it should be pointed out that the absorption rate is affected by a combination of reaction rates and mass transfer rates. The overall absorption performance is governed by the effect of reaction kinetics on mass transfer rates, rather than the absolute kinetic rates.

The overall rate of absorption will severely influence the assessment of process performance, especially energy expenditure. Darde et al. (2010) applied the extended UNIQUAC model to simulate the stripper, which was used to regenerate the NH_3 absorbent, and reported that the regeneration energy was 2.1 GJ/ton-CO_2. Subsequently, the abatement duty of controlling ammonia slip was reported (Darde et al., 2012a) to be ca. 0.2–0.4 GJ/ton-CO_2 by using 7.8 wt% NH_3 absorbent. The overall energy consumption required with 7.8 wt% NH_3 absorbent was lower than that required with 30 wt% MEA absorbent, which was reported as 4 GJ/ton-CO_2 (Alie et al., 2005). Mathias et al. (2010) simulated the CAP using the equilibrium model of Aspen Plus with 26 wt% NH_3 absorbent and reported that the stripper duty was 2.3 GJ/ton-CO_2 and the NH_3 abatement duty was 2.4 GJ/ton-CO_2, which is significantly larger than the results reported by Darde et al. (2012a) due to the high concentration of ammonia used. Versteeg and Rubin (2011) also used Aspen Plus to simulate the CAP model using an NH_3 concentration of 14.4 wt% in order to evaluate the power penalty and the capture cost. The respective power penalties obtained by 30 wt% MEA and CAP are 30.4% and 28.6%. Furthermore, the differences between the total capital cost and the operating cost per year for the two processes are approximately 5% and 1%, respectively. They concluded that the absorber cooling requirements and the ammonia cleanup system significantly increased the energy loads and capital costs for the CAP. However, in the aforementioned CAP simulations, the reaction kinetics and the vapor-to-liquid mass transfer rate were not taken into consideration.

Because CO_2 absorption is limited by mass transfer in the liquid phase, the evaluation results achieved with the equilibrium model of Aspen Plus are overly optimistic (Mathias et al., 2010). Niu et al. (2012) conducted the CO_2 absorption tests on the laboratory scale and the experimental data were used to develop a process model according to the rate-based model of Aspen Plus. They concluded that the optimal operating conditions involve the use of an NH_3 concentration in the absorbent of 5–7 wt% and a CO_2-lean loading of 0.12–0.15, whereas over 90% CO_2 removal efficiency could be achieved. Furthermore, increasing the NH_3 concentration beyond 7 wt% had minimal effect on the removal efficiency, whereas the ammonia slip increased significantly. Zhang and Guo (2013) developed a large-scale CO_2 capture process based on aqueous ammonia solution using the rate-based model of Aspen Plus, and validated the absorption model using the data from pilot trials at the Munmorah Power Station (Yu et al., 2011). They evaluated a 500 MW coal-fired power plant, where the amount of CO_2 captured can reach 1.8 million tons per year, and found that the required diameter and packed height of the absorber were 40 and 72 m,