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A low energy aqueous ammonia CO_2 capture process

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Abstract

The most pressing challenges regarding the use of ammonia for CO_2 capture are the precipitation limitation and the energy penalty of solvent regeneration. Precipitation-free operation is a vital task since solids may cause the shutdown of the plant. Precipitation and slurry formation can be avoided by increasing temperature and L/G ratio but this leads to higher heat consumption, jeopardizing the economic feasibility.
Here we developed, investigated, and optimized a novel CO_2 capture process design using aqueous ammonia as solvent. The proposed configuration replaces the traditional stripper for solvent based CO_2 capture with a thermal decomposition reactor. The overall energy penalty is reduced at the expense of introducing a solid handling section which consists of a saturation reactor, a crystallizer and a belt filter.
The feasibility of the present approach is demonstrated by simulation. Flow-sheet calculations are performed in Aspen Plus using the extended UNIQUAC thermodynamic model for vapor-liquid-solid equilibria and for thermal properties calculation of the CO_2-NH_3-H_2O system.
The simulation results show that the specific regeneration duty of the novel capture alternative is comparable with existing aqueous ammonia CO_2 capture processes. Moreover, the thermal reactor can operate at 1 bar and 86 °C, therefore the NH_3 regeneration temperature is reduced by approximately 50 °C. The integration of low- and mid- temperature waste heat becomes possible which can greatly improve the economics of the process. The present capture alternative is especially convenient for power plants but is also beneficial for the cement, steel and aluminum industry.
Special attention is given to the ammonia slip prediction. The calculations substantiate that the slip above the absorber is 0.1 mol % after washing with the rich solution and it reduces below 100 ppm by washing with low temperature water.

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Keywords: CO_2 post-combustion capture simulation and optimization; aquoeus ammonia; slurry and solid formation; low energy process;extended UNIQUAC

1. Introduction

The great paradox of the 21st century is to meet the increasing global demand for energy and products while simultaneously mitigating the climate change [1]. If both these criteria are to be met, carbon capture and storage is an imperative technology for sustainable energy infrastructure development.

Several capture technologies exist at varying levels of maturity. Post-combustion capture is a so-called “end-of-pipe” technology which is suitable for various processes in power plants, steel industry, cement production, and bio-chemical industry. It is particularly attractive since it can be retrofitted to existing plants.

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Today there is a potential for innovative solvent designs and process configurations. One promising solvent for the CO_2 capture process is ammonia, due to its low cost, limited degradation and fast reaction kinetics. The technical feasibility of the aqueous ammonia CO_2 capture process is proven, meaning that the CO_2 capture percentage can be maintained at 90% with CO_2 purity over 95%. The successful commercialization of ammonia based carbon capture technology narrows down to the economic feasibility optimization. For this, the energy consumption of the regeneration step needs to be lowered. In addition, some technical issues including ammonia slip, precipitation and slurry formation need to be resolved before the technology can be deployed [2].

Several research groups are developing different variants of the of the aqueous ammonia CO_2 capture process to replace the traditional amine-based technology. One of the leading approaches is the chilled ammonia process (CAP) developed by Alstom Power Company [3]. The CAP relies on the exhaust gas treatment with high concentration (~28 wt%) ammonia solution at very low temperatures, preferably below 10 °C. The main issue of the CAP process is the energy consumption associated with cooling and the high ammonia slip and washing plus regeneration of the wash water. It might also suffer from solid precipitation, depending on the process condition. One way to reduce the ammonia slip is to decrease the lean solution ammonia content, for example the CSIRO process which uses ~6wt% NH_3 solution [4,5]. However, low NH_3 concentration results in higher energy consumption for CO_2 stripping respectively ammonia regeneration and much higher electricity cost for circulating the liquid. Moreover, due to high loading and low temperature of the rich solution, precipitation and solid handling is a key challenge for the CSIRO process [2]. A unique CO_2 capture process, using low concentration (~10 wt%) aqueous ammonia solution is the RIST process. This process is optimized for the iron and steel industry. Although it is an energy intensive process, Rhee et al. [6] presented the economic feasibility of the process by recovering low- and mid-temperature waste heat from the boiler stack at 140 – 150 ºC.

The design of the above ammonia processes is based on a standard absorption and desorption column. The difference between them relies in the operational conditions, such as different ammonia concentration of the lean solution, the operating temperature range of the absorber and regenerator, and different pressures. All of these technologies face some common technical issues related to the volatile nature of ammonia, the energy intensiveness of gas cooling and solvent regeneration, and finally the risk of clogging of the equipment due to solid precipitation.

In the present work a low energy CO_2 capture process using aqueous ammonia is developed and investigated. This novel alternative addresses the main issues of existing process configurations. The overall energy penalty is reduced at the expense of introducing a solid handling section which consists of a saturation reactor, a crystallizer and a filter. In contrast with the traditional solvent based CO_2 capture, the present configuration is centered on thermal decomposition of a solid.

The feasibility of the process is evaluated by simulation. The plant model is based on the extended UNIQUAC thermodynamic framework which is able to accurately predict vapor-liquid-solid equilibrium (VLSE) in the CO_2-NH_3-H_2O system. This analysis reveals that the regeneration duty is comparable with existing aqueous ammonia capture processes. Special attention is given to the NH_3 slip evaluation. An ethyl-glycol based heat pump is used to maintain the temperature at 2 °C. Simulation suggests that the NH_3 slip is below 100 ppm by washing with CO_2-loaded solution and water, and down to 1 ppm by washing with acid solution.

2. Extended UNIQUAC thermodynamic model for NH_3-CO_2-H_2O system

The extended UNIQUAC thermodynamic model proposed by Thomsen and Rasmussen [7] and upgraded by Darde et al. [8] for NH_3-CO_2-H_2O system is applied in the present work to accurately describe the solid–liquid–vapor equilibria as well as the thermal properties. The phase equilibrium is calculated in a γ–ϕ approach coupled with equilibrium speciation reactions with solid precipitation. Therefore, liquid phase activity coefficients are calculated with the extended UNIQUAC model, and the gas phase fugacity coefficients are estimated with the Soave–Redlick–Kwong equation of state [9], solid phase are assumed to be pure.

Extended UNIQUAC is a Gibbs excess energy model with adjustable parameters to account for the system specific interactions between ions and molecules. These parameters have been found through data reduction by fitting to more than 3700 experimental data points from various types of experiments, such as vapor–liquid equilibrium, solid–liquid equilibrium, enthalpy measurements, speciation data and heat capacity [10]. A description of how parameter fitting is performed and the framework surrounding it is detailed by Thomsen [11]. This model has been implemented in Aspen Plus® commercial simulator using a user model interface, developed by Maribo-Mogensen [8].

The excellent behavior of the extended UNIQUAC model in the NH_3-CO_2-H_2O system for absorption and desorption was demonstrated by Darde et al. [8]. Furthermore, it was compared against the e-NRTL model implemented in Aspen Plus®. The analysis has revealed that the extended UNIQUAC model is valid for the whole loading range up to 150 °C. It performs better than e-NRTL for the system of interest. The study outlines that the partial pressure and solid formation is better estimated using the extended UNIQUAC model. It describes the solid-liquid-vapor equilibria, speciation and thermal properties of the system are well predicted using this model[10]. DTU's version of the extended UNIQUAC Aspen Plus user model is used in the present work for the novel ammonia capture process simulation.