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Numerical simulation of aqueous ammonia-based CO_2 absorption in a sprayer tower: An integrated model combining gas-liquid hydrodynamics and chemistry

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HIGHLIGHTS

- A model integrating hydrodynamics and chemistry of NH_3-CO_2-H_2O system is developed.
- CO_2 removal efficiency and NH_3 escape are studied experimentally and numerically.
- Model using Liu's kinetics presents a better agreement with the experiments.
- Distribution of gaseous species and temperature depends on gaseous velocity field.
- Alternation of CO_2 removal efficiency at 303 K is observed and explained.

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ABSTRACT

CO_2 capture in the spray tower using aqueous ammonia solution has been proved a potential approach for the post-combustion CO_2 capture. To improve the performance of spray absorption, a new three-dimensional numerical model integrating the complex gas-liquid hydrodynamics and the chemistry of the NH_3-CO_2-H_2O system has been proposed and developed in this paper. In the model, the gas-liquid hydrodynamics was simulated using the Euler-Lagrange method, while the chemistry of the NH_3-CO_2-H_2O system, including both thermodynamic and kinetic characteristics, was modeled using user-defined modules implemented in the CFD package. Moreover, a laboratory scale spray absorption system was built, and run under various operating conditions were carried out in order to validate the model developed. Subsequently, the predicted results using different kinetic models were compared with the experimental results. It was found the model using Liu's kinetics showed a better agreement with the experiments, especially when the liquid temperature and NH_3 concentration of solution vary. Furthermore, the validated model was used to study the fundamentals of spray absorption. It has been found that the gas temperature profile as well as gaseous CO_2 and NH_3 concentration profile depends on the velocity field, indicating the hydrodynamics should be considered when optimizing the design of reactor. It has also been found that the residence time of droplets increases with the increase of gas flow rate, leading to an increase in average droplets temperature and CO_2 loading, while the increase of ammonia concentration of solution could significantly improve the overall CO_2 mass transfer coefficient. When the solution temperature increases from 293 K to 303 K, the overall CO_2 mass transfer coefficient gradually increases, resulting in CO_2 removal efficiency improvement. However, when the solution temperature increases from 303 K to 313 K, the promotion of reverse reaction leads to the reduction of CO_2 removal efficiency. In addition, the increase of ammonia solution flow rate would mainly increase the interphase area and promote both CO_2 absorption and NH_3 escape. The model developed in this paper can be used to optimize the design and operation of such reactors in practice.

Abbreviations: CFD, computational fluid dynamics; CSIRO, Australia's commonwealth scientific and industrial research organization; E-NRTL, the electrolyte nonrandom two-liquid model; IPCC, the intergovernmental panel on climate change; RNG, the renormalization group theory

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1. Introduction

Research and development of CO_2 capture and storage (CCS) technologies are of great urgency and utmost importance for relieving environmental issues such as global warming, sea level rise, coastal region erosion, and abnormal climate [1–3]. According to a variety of reports, fossil-fuel combustion is a major contributor of anthropogenic CO_2 emissions, particularly coal combustion for electricity generation [4–7]. Therefore, over the past few decades, mitigating CO_2 emissions from thermal power plants have become popular topics and several technical options have been proposed, including post-combustion capture, pre-combustion capture, oxyfuel combustion, and chemical looping [8–11]. Among these alternatives, post-combustion capture (PCC) can be installed directly to existing plants, exhibiting an attractive economical advantage, which can also accelerate the implementation of CCS [12–17]. Previous studies on PCC widely used an aqueous monoethanolamine (MEA) solution as CO_2 absorbent [18–21]. However, the issues of high regeneration energy, easy degradation by SO_2 and O_2, and facilities corrosion, are always accompanied with the utilization of MEA [22]. As a promising alternative, an aqueous ammonia solution has been proposed due to the advantages of high capacity, low cost, less corrosion, low regeneration energy, no degradation in the presence of O_2 and SO_2, and the potential of removing CO_2, NO_x, and SO_2 simultaneously [23–25]. Consequently, ammonia-based CO_2 capture has been extensively studied in recent years. The thermodynamic data and models of NH_3-CO_2-H_2O system have been upgraded [26–28]. The kinetics of CO_2 absorption into ammonia aqueous solution has also been discussed thoroughly [29–38]. Pilot plants have been built to demonstrate the technical and economical feasibility of ammonia-based CO_2 capture by Alstom [39], Powerspan [40], and CSIRO [41,42], respectively. In the mean time, process simulation has been carried out to evaluate the performance of ammonia-based CO_2 capture [43–48]. Through these efforts, the technical feasibility of ammonia-based CO_2 capture has been successfully proven, which could achieve 90% CO_2 removal efficiency and 99% purity of product CO_2 [49]. However, special consideration should be paid to the issue of ammonia escape, which may result in ammonia loss and secondary pollution. That also hinders the industrial application of ammonia-based CO_2 capture [50]. For the worse, solid precipitation due to the reaction of escaped ammonia and CO_2 could cause plugging of pipes and valves, leading to a shutdown of the whole system [42].

Measures of installing additional washing system and cooling down the flue gas have been proposed to control ammonia escape [51,52]. However, these measures consume extra power and water, lowering the economical feasibility of ammonia-based CO_2 capture. Therefore, adding functional additives into the solution becomes another way of inhibiting ammonia escape. Various additives have been tested in labs, including hindered amines [53], alcohols [54], metal cations [55–57], and ionic liquids [58]. However, experimental results indicate that most additives have negative influence on CO_2 capture. Suitable additives are not yet available.

CO_2 absorption and ammonia escape are also influenced by reactor configuration and operating conditions. Thus, optimizing design of a reactor and its operating conditions can provide a potential solution to abovementioned issues. Over the past few years, ammonia-based CO_2 capture using packed columns has been studied extensively [59,60]. Especially, a rotating packed column has been proposed to intensify multiphase transportation [61]. On the contrary, there have been few attempts in utilizing the spray columns for ammonia-based CO_2 capture. Compared with the packed columns, the spray columns have a few advantages, such as low pressure drop, less susceptibility to corrosion, suitability for treating large volumes of flue gas, and greater tolerance to solid precipitation. These features have led to the broad industrial application of spray columns in desulfurization [62]. Kuntz et al. [63] compared the performance of a spray column with a packed column for CO_2 absorption into MEA solution. They found that the spray column was capable of removing CO_2 from gas mixture at a higher rate than that of the packed column. Javed et al. [64] investigated CO_2 absorption into NaOH solution in a high-intensity vortex spray scrubber. The experimental results showed imparting swirl in the gas flow can significantly enhance the overall mass transfer coefficient. Bandyopadhyay et al. [65] used a two-phase critical flow atomizer for CO_2 absorption into dilute NaOH solution, achieving a high CO_2 removal efficiency. Ma et al. [66] investigated various operating conditions on the volumetric overall mass transfer coefficient of ammonia-based CO_2 capture in a spray tower. Very recently, ammonia-based CO_2 capture in a vortex spray scrubber was theoretically and experimentally investigated by Zhao et al. [67]. The studies demonstrated a great potential of using spray columns in CO_2 capture. However, the focus of the studies was mainly concentrated on the influence of operating conditions on CO_2 removal efficiency or mass transfer coefficients. The fundamentals of spray absorption have not been fully understood.

An integrated model combining complex gas-liquid flow and chemical reaction as well as heat transfer is of benefit to comprehensively understand CO_2 absorption process and optimize design of a reactor. In previous studies, Kvamsdal et al. [68] and Lawal et al. [69] employed gPROMS to develop 1-D dynamic models for CO_2 absorption by MEA with the assumption of plug flow regime. Niu et al. [43] used a rate-based model implemented in Aspen Plus to simulate ammonia-based CO_2 capture in a packed column, in which only the axial flow was considered. These 1-D models simplified the complex gas-liquid flow which in fact plays a vital role in the absorption process. In recent studies, much emphasis has been placed on the complex hydrodynamics in absorbers. Niegodajew et al. [70] used the Euler-Euler multiphase models to consider gas-liquid flow in a random packed column. Chu et al. [71] employed the VOF model to consider the gas-liquid flow in a bubble column. For spray columns, Chen et al. [72,73] and Choi [74] theoretically investigated behavior of a single droplet in a spray tower. Zhao et al. [67] investigated the gas flow pattern in a vortex spray column using the RANS model without consideration of reactions. Clearly, there still lacks an integrated model for ammonia-based CO_2 capture in a spray column, limiting the development of ammonia-based CO_2 capture.

In the present work, the principal aim is to establish an integrated model combing the complex gas-liquid flow and the chemistry of the NH_3-CO_2-H_2O system. Both the processes of CO_2 absorption and ammonia slip were taken into consideration. Then, the predicted results were compared with the experimental data from a lab-scale spray column to valid the established model. Moreover, the influence of operating conditions on both CO_2 removal capture and ammonia slip was studied experimentally and theoretically. To reveal the fundamental mechanisms and understand the spray absorption, the validated model was employed to investigate the velocity, temperature and species distributions in the spray column. The understanding will benefit the development of ammonia-based CO_2 capture.

2. Physical models and mathematical formulations

The absorption of CO_2 in a sprayer tower is a typical two-phase flow consisting of continuous flue gas and dispersed liquid droplets. Therefore, the Euler-Lagrange method is adopted due to the advantages of rigorous description of particle-level phenomenon, droplet size distribution, and droplet-wall interaction [75]. The flue gas is treated as a continuum, described by the Eulerian approach, whereas the dispersed droplets are modeled by the Lagrangian approach. Interphase mass and heat transfer is realized via User-defined Functions coupled to the CFD code. Besides, the following assumptions should be made:

(1) The shape of the droplets remains spherical.
(2) Particle-particle interactions in the tower are neglected considering the volume fraction of the droplets is much lower than 10 vol% in the spray reactor except the domain very close to the nozzle.