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Modeling solid-liquid equilibrium of the NH_3-CO_2-SO_2-K_2SO_4-H_2O system and its application to combined capture of CO_2 and SO_2 using aqueous ammonia

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Abstract

It is of significance to intensively investigate the solid-liquid equilibrium of the NH_3-CO_2-SO_2-K_2SO_4-H_2O system to assess the novel aqueous NH_3 based CO_2 and SO_2 combined capture process proposed in our previous work. In this work, the solubility of K_2SO_4 in aqueous NH_3 at various temperatures, CO_2 and SO_2 loadings, and NH_3 concentrations was predicted using Aspen Plus with a developed thermodynamic model package, which can accurately simulate the vapor-liquid equilibrium and the solid-liquid equilibrium of the combined CO_2 and SO_2 capture system. The results indicate that the solubility of K_2SO_4 increases with temperature and CO_2 loading, but decreases with NH_3 concentration. The precipitation starting point of K_2SO_4 shift to higher temperature with the increasing of SO_2 loading and NH_3 concentration. It is favorable to collect K_2SO_4 precipitates at lower CO_2 loading and temperature, and higher SO_2 loading and NH_3 concentration.

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Keywords: Solid-liquid equilibrium; combined capture; CO_2; SO_2; K_2SO_4; Aspen Plus;

1. Introduction

Carbon dioxide (CO_2) and sulfur dioxide (SO_2) are two of the main impurities from coal-fired power stations, which could be captured by flue-gas desulfurization (FGD) system and post-combustion CO_2 capture (PCC) facility [1,2,3]. However, both FGD and PCC systems are very expensive, and cause significant reduction of the power

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station efficiency [4]. Aqueous ammonia (NH_3), as an alternative chemical absorbent, has greatly potential to efficiently capture CO_2 and SO_2 together [5,6]. A novel post-combustion CO_2 and SO_2 combined capture process (without typical FGD system) using aqueous ammonia was proposed in our previous work, as shown in Figure 1, in that both CO_2 and SO_2 can be captured in one absorber, the CO_2 related content in CO_2 and SO_2 enriched solvent can be thermally regenerated in a stripper, and the SO_2 related content can be removed from the rich solvent by integrated sulfite forced oxidation and sulfate precipitation process [7].

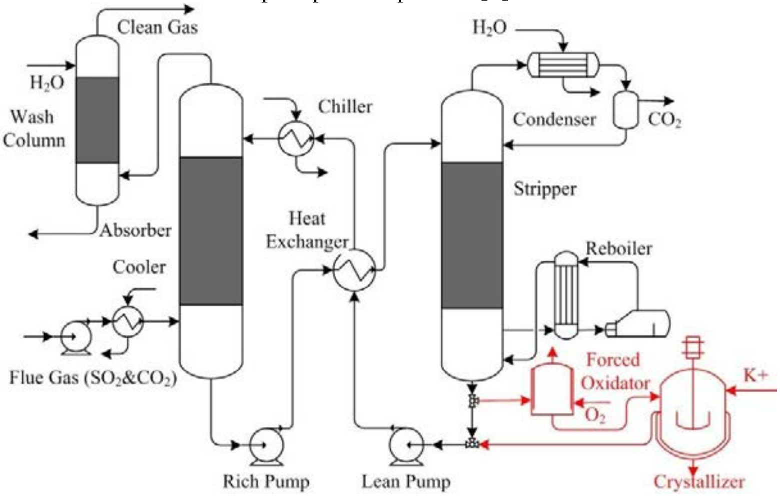

Fig. 1. Proposed combined CO_2 and SO_2 capture process without FGD system.

In our previous work, the potassium additives (e.g. K_2SO_4) are proved and selected as the media for SO_2 content removal in the combined capture process [5]. However, the detailed study on the solubility of potassium salts in the CO_2 and SO_2 loaded aqueous NH_3 solvent is lacking in literature. Hence, it is of significance to intensively investigate the solid-liquid equilibrium of the NH_3-CO_2-SO_2-K_2SO_4-H_2O system and confirm the feasibility of its application in the combined capture of CO_2 and SO_2 using aqueous NH_3.

In this work, a solid-liquid equilibrium model was built using the Aspen Plus with our developed thermodynamic model package for combined CO_2 and SO_2 capture. The package can accurately calculate the solubility of K_2SO_4, K_2SO_3, K_2CO_3, $KHCO_3$, $(NH_4)_2SO_4$, $(NH_4)_2SO_3$, and NH_4HCO_3 between 0 to 100 ºC, as shown in Figure 2.

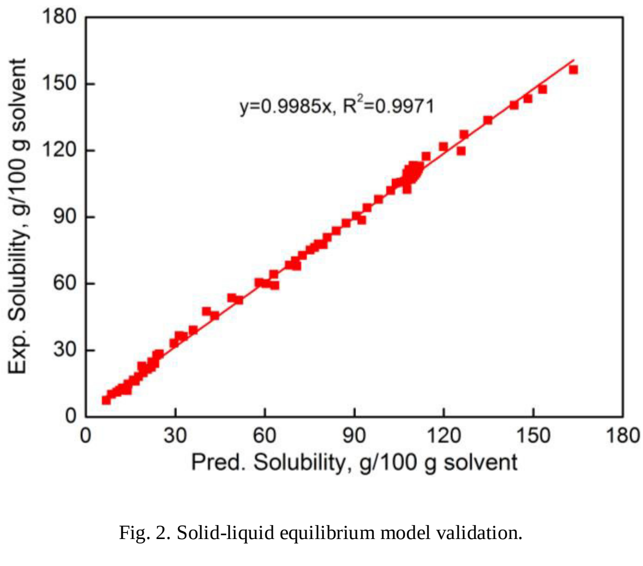

Fig. 2. Solid-liquid equilibrium model validation.