



Gas–liquid absorption reaction between $(\text{NH}_4)_2\text{SO}_3$ solution and SO_2 for ammonia-based wet flue gas desulfurization

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ABSTRACT

In order to investigate the characteristics of the reaction between ammonium sulfite, the main desulfurizing solution, and the flue-gas-contained sulfur dioxide during the process of ammonia-based WFGD (wet flue gas desulfurization) in a power plant, the gas–liquid absorption reaction between sulfur dioxide and an ammonium sulfite solution was studied in a stirred tank reactor. The experimental results indicate that the absorption of sulfur dioxide is controlled by both the gas- and liquid-films when the ammonium sulfite concentration is lower than 0.05 mol/L, and mainly by the gas-film at higher concentrations. In the latter case, the reaction rates are found to be zero-order with respect to the concentration of ammonium sulfite. The absorption rates of sulfur dioxide increase as the concentration of sulfur dioxide in inlet gas and the temperature increase. The reaction rate is of 0.6th-order with respect to the concentration of sulfur dioxide.

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1. Introduction

The environmental pollution caused by energy consumption is a globe issue [1–4]. Other than the emission of CO_2 and NO_x , SO_2 caused by fossil fuel combustion is an important factor leading to environmental pollution [5–8]. To control the sulfur dioxide emission, various flue gas desulfurization (FGD) processes have been developed and applied worldwide. Among them, wet flue gas desulfurization (WFGD) is the main technology [9]. In recent years, ammonia-based wet flue gas desulfurization (Am-based WFGD) has drawn increasing attention in China [10] because of its lower investment, higher desulfurization efficiency, no secondary pollution, and useful byproducts.

In the Am-based WFGD process, $(\text{NH}_4)_2\text{SO}_3$ and NH_4HSO_3 coexist in the desulfurization liquid without free NH_3 and SO_2 . Ammonium sulfite is the key compound removing sulfur dioxide from flue gas [11]. Ammonia added to the system can adjust the pH value and $(\text{NH}_4)_2\text{SO}_3$ concentration in aqueous solution. Therefore, it provides basic data for the process of investigating the role of the gas–liquid reaction between SO_2 and the $(\text{NH}_4)_2\text{SO}_3$ solution. However, detailed information on the kinetics of the heterogeneous reaction between sulfur dioxide and ammonium sulfite solution is not readily available in the literature. Most of the similar studies

focus on calcium-based desulfurization processes [12–14]. The above reasons necessitate further studies on the gas–liquid reaction between sulfur dioxide and ammonium sulfite solution.

The role of mass transfer and the absorption reaction can be effectively determined using a stirred tank reactor. Uchida et al. [13] studied the absorption of lower concentration sulfur dioxide into limestone slurries in a stirred tank reactor. The absorption rates of sulfur dioxide were measured under various conditions. Through analyzing the experimental results, Uchida concluded that the limestone dissolution process seems to play a very important role in the absorption. Long et al. [15] performed a series of experiments on the chemical absorption of nitric oxide into the $\text{Co}(\text{NH}_3)_6^{2+}$ solution in a stirred cell under anaerobic and aerobic conditions separately, and discussed the impacts of nitric oxide on the absorption rates.

In this work, the absorption reaction of sulfur dioxide into ammonium sulfite solution was studied in a stirred reactor. The aim of this study is to investigate the characteristics of gas–liquid absorption reaction between sulfur dioxide and ammonium sulfite solution.

2. Theories

According to the theory of absorption with chemical reaction, the reaction between SO_2 and $(\text{NH}_4)_2\text{SO}_3$ is likely to conform to the instantaneous reaction regime. Stirred tank reactors can be used to investigate the characteristics of fast or instantaneous reactions. In a stirred tank, there is a gas–liquid interface which is

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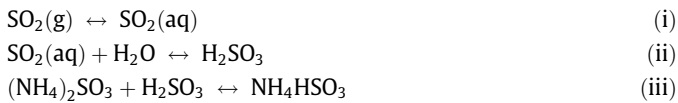
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Nomenclature

A	interfacial area, m^2	K_G	overall mass transfer coefficient using partial pressure as driving force, $\text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$
c_{s1}	SO_2 concentration of inlet gas, kg m^{-3}	N_s	absorption rate of SO_2 , $\text{mol m}^{-2} \text{s}^{-1}$
c_{s2}	SO_2 concentration of outlet gas, kg m^{-3}	p_s	partial pressure of SO_2 in bulk of gas, Pa
c_i	SO_2 concentration at interface, mol m^{-3}	p_i	partial pressure of SO_2 at interface, Pa
c_{SL}	SO_2 concentration in bulk of liquid, mol m^{-3}	Q_s	gas flow rate, $\text{m}^3 \text{s}^{-1}$
c_{NL}	$(\text{NH}_4)_2\text{SO}_3$ concentration in bulk of liquid, mol m^{-3}	R	gas constant, $\text{m}^3 \text{Pa mol}^{-1} \text{K}^{-1}$
D_{NL}	diffusivity of $(\text{NH}_4)_2\text{SO}_3$ in water, $\text{m}^2 \text{s}^{-1}$	T	temperature, K
D_{SL}	diffusivity of SO_2 in water, $\text{m}^2 \text{s}^{-1}$	γ_L	conversion coefficient in liquid-film
D_{CL}	diffusivity of CO_2 in water, $\text{m}^2 \text{s}^{-1}$	M	molar weight of SO_2 , kg mol^{-1}
E	enhancement factor	m	order of reaction with respect to concentration of SO_2 in bulk gas
E_∞	enhancement factor when reaction is instantaneous	n	order of reaction with respect to concentration of $(\text{NH}_4)_2\text{SO}_3$
H	Henry's law constant, $\text{Pa m}^3 \text{mol}^{-1}$		
k_G	gas phase mass transfer coefficient, $\text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$		
k_L	liquid phase mass transfer coefficient, m s^{-1}		

approximately planar. The mass transfer and absorption reaction are carried out between the gas- and liquid-phases through this interface. According to the film theory, there is a stagnant film on both sides of the gas-liquid interface. Transfer resistance exists in the film, and the mass transfer within the film is achieved by molecular diffusion. Sulfur dioxide must spread from the gas phase to the liquid-film, and then into the bulk of liquid. The reaction occurs mainly in the liquid-film, while the bulk liquid is at equilibrium.

The following reactions will mainly occur during the absorption of SO_2 into $(\text{NH}_4)_2\text{SO}_3$ solution when no oxide is present in the environment [16]:



In certain circumstances, a stirred tank reactor can be considered to be in the completely mixed state [17]. In this case, the gas-phase component of the outlet gas of the tank can be regarded approximately as that of the inner gas of the tank.

In order to study the reactions under different experimental conditions, the absorption rates of SO_2 by the material balance calculation for the gas phase is given by [18]:

$$N_s = Q_s \cdot (c_{s1} - c_{s2}) / AM \quad (1)$$

Based on the two-film model, the mass transfer equation of SO_2 into $(\text{NH}_4)_2\text{SO}_3$ solution is [18]:

$$N_s = k_G(p_s - p_i) = Ek_L(c_i - c_{SL}) \quad (2)$$

Combining the above with the phase balance in the gas-liquid interface, $c_i = p_i/H$, the total mass transfer equation is expressed by the gas phase impetus as follows:

$$N_s = \left(p_s - \frac{c_{SL}}{H} \right) / \left(\frac{1}{k_G} + \frac{1}{EHk_L} \right) \quad (3)$$

Assuming that the reaction (iii) is an m , n th-order reaction, and $c_{SL} = 0$ in the interface, the enhancement factor E can be approximated by [19]:

$$E = \frac{\gamma_L [(E_\infty - E) / (E_\infty - 1)]^{n/2}}{\tanh\{\gamma_L [(E_\infty - E) / (E_\infty - 1)]^{n/2}\}} \quad (4)$$

where E_∞ and γ_L can be obtained by:

$$E_\infty = 1 + D_{NL}c_{NL} / (D_{L,\text{SO}_2}c_{si}) \quad (5)$$

$$\gamma_L = \frac{1}{k_L} \left[\frac{2}{m+1} k_{m,n} D_{L,\text{SO}_2} c_i^{m-1} c_{NL}^n \right]^{1/2} \quad (6)$$

For a fast pseudo m , n th-order reaction, $1 \ll \gamma_L \ll E_\infty$. In this case, $E = \gamma_L$. Substituting these values, the absorption rate of SO_2 can be written as:

$$N_s = \frac{p_s}{\frac{1}{k_G} + \frac{1}{H \sqrt{\frac{2}{m+1} k_{m,n} D_{L,\text{SO}_2} c_i^{m-1} c_{NL}^n}}} \quad (7)$$

3. Experimental

A schematic diagram of the experimental system is shown in Fig. 1. All of the experiments on the absorption of SO_2 into $(\text{NH}_4)_2\text{SO}_3$ solution were carried out in a stirred tank reactor. The main part of the tank consisted of an organic glass vessel of 100 mm diameter and 160 mm height, equipped inside with 4 baffles of 8 mm width. The liquid volume in the tank was 800 cm^3 . Two 4-blade impellers were used for mixing the gas and the liquid at the same speed. The rotational speed of the impellers was 260 rpm. The tank was immersed in a water bath to maintain the temperature of the gas and the liquid within $\pm 0.5 \text{ }^\circ\text{C}$.

The $(\text{NH}_4)_2\text{SO}_3$ solution (28%) is a product of SCRC and was diluted with distilled water to get the desired concentration. The concentration of the $(\text{NH}_4)_2\text{SO}_3$ solution was analyzed by iodimetric technique.

SO_2 (5000 ppm in N_2 , Xinshiji) was supplied from a cylinder and was diluted with N_2 (>99.99%, Xinshiji) from another cylinder. The two kinds of gases went into a gas mixer to form the simulated flue gas with a required concentration. The simulated flue gas was then fed into the stirred tank reactor and reacted with the $(\text{NH}_4)_2\text{SO}_3$ solution. When the experimental process was stable, the outlet gas concentration was measured [20]. The gas flow rates were controlled by mass flow meters purchased from SevenStar Huachuang Co., Ltd. China. A SO_2 analyzer, Testo 350 pro from Testo Instrumental Trading (Shanghai) Co., Ltd. was used to measure the concentration of the absorbed gas.

4. Results and discussions

4.1. Mass transfer coefficient of the gas- and liquid-films

The gas- and liquid-film mass transfer coefficient was measured based on the methods of Shi et al. [21], Nagel et al. [22] and Chu et al. [23]. The range of the experimental temperature was $20\text{--}50 \text{ }^\circ\text{C}$.

The knowledge of diffusivity and physical solubility of SO_2 and CO_2 were necessary for the calculation in this work. The solubility

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