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SO_2 removal by seawater in a spray tower: Experimental study and mathematical modeling

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ABSTRACT

In this article, the reactive absorption of SO_2 by seawater is studied in a spray tower experimentally and mathematically. The liquid film formation on the tower wall is implemented in the model and measured experimentally at different operating conditions. The effect of liquid to gas flow rate, initial SO_2 concentration in gas phase and initial gas temperature on SO_2 removal efficiency is examined. Regarding the importance of liquid droplets hydrodynamics and its effect on the performance of the equipment, the required differential equations for predicting the trajectory and local velocity of droplets are also developed based on the nozzle and spray characteristics and solved simultaneously with other governing equations. In order to survey the effect of nozzle type on removal efficiency, two different types of nozzles are examined. Semi-empirical correlations are proposed for two different nozzles by using experimental data and droplets hydrodynamics model, to predict the amount and the variation of liquid film mass flow rate on the spray tower wall. Results indicate that neglecting the liquid film formation leads to an average of 23% error in predicting the removal efficiency when nozzle type 1 is used, while the calculated error of model by considering the film formation is reduced to 4%. By implementation of droplets hydrodynamics model and applying a modified thermodynamics model for predicting the behavior of the existing chemical reactions, the capability of the spray tower model in predicting the SO_2 removal efficiency is enhanced.

1. Introduction

Recently, the international committees have adopted several restrictions for depletion of the released SO_2 from industries (Vidal et al., 2007). In the past few decades, studies have focused dramatically on the flue gas desulphurization (FGD) process. Meanwhile, different equipment such as venturi scrubbers, spray towers, tray towers, packed beds (Bandyopadhyay and Biswas, 2006, 2008; Gamisans et al., 2002), and membrane technology (Sun et al., 2008) have been studied for FGD process in a vast range.

The most common method for FGD is scrubbing the polluted gas through alkali solvents like urea, dilute NaOH, limestone slurry, NaCl solution, water, and seawater (Barbooti et al., 2011; Bokotko et al., 2005; Jeong and Kim, 1997). According to searches in this respect, a few studies were conducted on using seawater as an alkali absorbent for SO_2 removal. Sun et al. (2008) studied SO_2 absorption by seawater in a hollow fiber membrane contactor and found that the mass transfer coefficient in seawater is about double the mass transfer coefficient in the NaOH solution content with a pH of 8.35.

Abbreviations: FGD, flue gas desulfurization; ppm, parts per million by volume; ppt, parts per trillion.

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Nomenclature

D	diameter of tower (m)
d	droplet diameter (m)
C_{pg}	heat capacity of gas (J/kg K)
C_{pl}	heat capacity of liquid (J/kg K)
C_{pv}	heat capacity of vapor (J/kg K)
Q_g	flow rate of gas (m^3/s)
Q_l	total flow rate of liquid
Q_d	flow rate of droplet
Q_f	flow rate of liquid film
H_{SO_2}	Henry's constant
h	heat transfer coefficient (W/m^2 K)
I	ionic strength
k_{wd}	mass transfer coefficient for water between gas and droplet phase (mol/m^2 s)
K_{wf}	mass transfer coefficient for water between gas and droplet phase (mol/m^2 s)
K_{gd}	mass transfer coefficient for SO_2 between gas and droplet (mol/m^2 s)
K_{gf}	mass transfer coefficient for SO_2 between gas and droplet (mol/m^2 s)
k_i	thermodynamic equilibrium constant
M_g	molecular weight of gas (kg/kmol)
V_{re}	relative velocity of droplet
M_l	molecular weight of liquid (kg/kmol)
m_i	concentration of ion
N_{sd}	molar flax of SO_2 between gas and droplet (mol/m^2 s)
N_{sf}	molar flax of SO_2 between gas and film (mol/m^2 s)
N_{wd}	molar flax of water vapor from droplet (mol/m^2 s)
N_{wf}	molar flax of water vapor from liquid film (mol/m^2 s)
y_s	SO_2 concentration at gas bulk
y_w	water concentration at gas bulk
y_{wi}	water concentration at gas–liquid interfacial
Z_i	ion charge
C_D	drag coefficient
T_0	reference temperature
T_d	droplet temperature
T_f	film temperature

Greek letters

γ_i	activity coefficient for ion
ΔH_r	heat of SO_2 solution in seawater (J/mol)
ρ_g	density of gas (kg/m^3)
ρ_l	density of liquid (kg/m^3)

Al-Enezi et al. (2001) measured the solubility of SO_2 in the Persian Gulf seawater through a semi-continuous system. Rodriguez-Sevilla (2004) developed a thermodynamic model for SO_2 solubility in seawater as a function of pH and the temperature of seawater (278.15–318.15 K). They used an extended version of the Debye–Huckel theory (1972) and the Pitzer ion-interaction model (Bromley, 1972; Pitzer, 1991; Rodriguez-Sevilla, 2004). They found that the solubility of SO_2 in seawater is within 20–60% and 6–30% higher than its solubility in distilled water and NaCl solution of similar ionic strength, respectively. Zhang et al. (2011) investigated the effect of additives on seawater FGD by introducing different concentrations and types of the selected additives. Javed et al. (2006) conducted an experimental study to investigate the effect of imparting swirl to the axial gas flow on mass transfer coefficient through a spray tower. They developed a correlation for gas phase mass transfer coefficient as a function of the gas flow rate and initial droplet SMD[1]. Their method was based on the assumption of constant thickness of liquid film. Bandyopadhyay and Biswas (2008) carried out an experimental study to identify the important factors affecting on SO_2 absorption in a spray tower and survey of liquid flow rate. Their founding revealed that in liquid to gas ratio of 3 (L/m^3) and pH of 11.7, the removal efficiency reaches 100% for inlet gas with SO_2 concentration in range of 500–1500 ppm_v. A model based on the penetration theory was developed to calculate the dynamic absorption rate of sulfur dioxide by limestone slurry in a spray tower by Brogren and Karlsson (1997). In this model, instantaneous equilibrium reactions and reaction with finite rate were considered. Bandyopadhyay and Biswas (2007) presented a simple realistic model in order to investigate the effectiveness parameters such as the size and velocity of droplet, superficial gas velocity, liquid flow rate and tower height on the performance of a spray absorption tower. Besides, the removal efficiency was strongly affected by spray tower hydrodynamics, flow rates and dimensions. Gao et al. (2008) proposed a CFD model for SO_2 absorption by limestone slurry in a spray scrubber. The gas and liquid temperatures were adjusted at 298 K in this work. Besides, they overlooked the liquid film formation onto the tower wall and applied Rosin–Rammler rule for size distribution of initial droplets. Marocco (2010) developed a CFD model for prediction of removal efficiency close to the nozzle zone at isothermal condition, where the interaction between flue gas and slurry is high in spray column. Zhuang et al. (2015) developed a CFD model for NO_2 wet absorption including the chemical absorption by sodium sulfite in a spray tower.

Rahimi et al. (2002) developed a mathematical model to study simultaneous heat and mass transfer in hot gas spray system. Downing (1966) correlations were used to calculate heat and mass transfer coefficients. Bozorgi et al. (2006) developed a mathematical model by consideration of liquid film formation to study the performance of a spray scrubber of an industrial ammonium nitrate plant. Their results showed that the assumption of terminal velocity for the motion of droplets, especially for smaller ones is a rough assumption. Keshavarz et al. (2008) showed that the importance of the consideration of liquid film formation in a spray tower model depends on solubility of component in gas phase. Their model showed that neglecting the liquid film formation causes a smaller error for pollutant with low solubility. In order to predict absorption height in a spray tower, Zhu et al. (2014) investigated SO_2 absorption by limestone–gypsum. They revealed that absorption height declines with increasing liquid–gas ratio and pH value of the slurry, and increases with increasing droplet diameter, gas flow rate, gas temperature, inlet SO_2 concentration and absorption efficiency, respectively.

Vidal et al. (2007) carried out an experimental study on FGD by use of seawater in a catalytic packed tower. They worked in the range of liquid to gas ratios of 4–15 (L/m^3) and used active carbon in packed structure. They showed that a 47% reduction in seawater flow rate could be obtained in the catalytic packed tower relative to conventionally spray tower.

[1] Sauter mean diameter.