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Research Paper

The effect of solid particles on the evaporation and crystallization processes of the desulfurization wastewater droplet

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HIGHLIGHTS

- TGA-DSC and SEM methods were used to investigate the evaporation and crystallization processes.
- An investigation of the factors of the Peclet number was conducted to help measure the crystallization process.
- The crystallization rate increased with the increase of the mass of the fly ash, but there was an top limit.
- Unlike Al_2O_3, particles of SiO_2, $CaCO_3$, Fe_2O_3, Fe_3O_4 and fly ash could increase the crystallization rate.
- The adding fly ash dominated the change of the Peclet number when the volume of the droplet changed.

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ABSTRACT

In this study, the thermogravimetric analyzer and the differential scanning calorimetry measurements of the evaporation and crystallization rates of the desulfurization wastewater droplet were conducted. During the measurements, the surface of the droplet was adding different mass of fly ash and adding different species of solid particles, and the volume of the droplet is changing when adding fixed mass of fly ash. The surface of the crystals after evaporation was observed by a scanning electron microscope. A theoretical model analyzing each specific parameter, like the heterogeneous nucleation adsorption sites of density and the radius of the droplet, of the Peclet number was built. The change of the Pe number has the same trend with the experimental results. The experimental results showed that the evaporation rate almost kept in the same level with or without particles. From 1.5 to 3.5 mg fly ash added, higher mass of fly ash on the surface of the 2.0 μL droplet could decrease the crystallization period from 1.2 to 0.95 min and improve the porosity of the crystals, but there was a top limit (crystallization period 0.95 min for fly ash 2.5 mg) for the controlling volume droplet. Particles of SiO_2, $CaCO_3$, Fe_2O_3, Fe_3O_4 and fly ash could increase the crystallization rate, but Al_2O_3 could inhibit the nucleation process and increased the crystallization time from 1.2 min (pure wastewater) to 1.25 min. When the volume of the droplet is changing from 0.5 to 2.5 μL, the adding of fly ash dominated the change of the Pe number. The crystallization period increased from 0.9 to 1.15 min and the porosity also increased dramatically.

1. Introduction

The wet scrubber technology has been the dominated method for the desulfurization of the flue gas in the coal-fired power plants. Notably, to 2014 in China, wet scrubber technology [1] had been used in the over 80% of all the flue gas desulfurization projects, and in the USA, 69% of the 108 coal-fired power plants in the USA will adopt the wet scrubber technology by 2025 [2]. This technology could decrease the emission of the sulfur dioxide, but it also could bring the problems of water pollution because there are many species of ions in it, like the acid ions (chloride ions and sulfate ion) and heavy metal ions (mercury (Hg), selenium (Se) and arsenic (As)). These ions are very difficult to separate, and also, the separation project needs huge investment. Therefore, an effective way to deal with the desulfurization wastewater could really help reduce the water pollution from the coal-fired plants.

Traditionally, to deal with the desulfurization wastewater from the coal-fired power plants, some sorts of chemicals [3–6] were added into the wastewater and the precipitation was separated. In the previous works, Guan et al. [3] were focused on removing manganese (Mn) and Zinc (Zn) ions from the FGD wastewater.

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Nomenclature

A	frequence coefficient
B	constant coefficient
Cp	specific heat [J/(kg·K)]
c_{solute}	concentration [g/m^3]
D	diffusion coefficient [cm^2/s]
d	diameter of the droplet [mm]
E	energy [kJ/mol]
K	evaporation coefficient
G	activation energy [kJ/mol]
J	nucleation rate [mg/s]
k	reaction coefficient
k_B	the Boltzmann constant
L	latent heat [kJ/mol]
l	characteristic length [mm]
M	molar mass [g/mol]
$\dot{m}$	evaporation rate [mg/s]
N	number of the absorption sites
Pe	the Peclet number
P	pressure [Pa]
Q	heat [J]
r	radius of the droplet [mm]
S	degree of supersaturation [%]
T	temperature [°C]
u	velocity of the convection [mm/s]
v	molecular volume [nm^3]
x	pressure fraction [%]
Y	mass fraction [%]
μ	liquid viscosity [Pa·s]
λ	conduction coefficient [W/(m·K)]
τ	time [s]
ρ	density [g/m^3]
π	circumference ratio
α	characteristic diffusion coefficient [mm^2/s]
σ	interfacial tension [mN/m]

Subscript

b	boiling point
def	surface diffusion
des	desorption
e	equilibrium condition
l	liquid phase
m	mixed phases
max	maximum condition
p	nucleation particle
r	radiation condition
s	surface condition
total	total gas phase
vap	vapor phase
Δ	difference
∞	far field of ambient air
0	initial condition
1	heterogeneous condition

Also, in this way, thirteen selenium species from FGD wastewater were separated by Petrov et al. [4]. They detected the possibility of the occurrence of selenosulfate and selenocyanate using anion-exchange chromatography coupled to inductively coupled plasma mass spectrometry. Huang et al. [5] were mostly concentrated on removing selenium (Se), mercury (Hg) and nitrate (NO_3^-) from desulfurization wastewater. Ohki et al. [6] were focused on removing boron and mercury from simulated water using various absorbents.

Unlike the traditional method, a plenty of more environment-friendly and investment-saving projects were designed to solve the wastewater problem. An important one of them is zero liquid discharged (ZLD) system which was firstly introduced by Shaw [7]. An evaporated which was used to evaporate the water from the wastewater and to increase the concentration of the solutes, and a crystallizer which separated the residual wastewater were designed in this system. Based on Shaw's work, advanced ZLD systems [8] were designed. In order to deal with more heavy metal in the wastewater and achieve higher water recovery, a membrane system and a reverse osmosis system were assembled. However, in these designs [7,8], additional energy was supported to evaporate the wastewater. Based on the energy saving, Ran and Zhang [9] designed a ZLD system which sprayed the wastewater into the gas flue in the end part of the boiler and evaporated the wastewater with the residual heat of the flue gas. The residual particles from the evaporated droplets would be collected by the electrostatic precipitator.

According to Ran and Zhang's work [9], the evaporation is an important process which affects the life time directly. Up to now, there have been lots of studies [10–12] reported about this field. Hartfield and Farrell [10] studied the evaporation process of the free falling hydrocarbon droplet in the high pressure condition. They analyzed the effect of the gas pressure and the gas temperature on the change of the diameter during the evaporation. Yin [11] studied the evaporation characteristics of moving mono-component liquid droplets and built a model for transient heating and evaporation of moving droplet. Wu et al. [12] investigated the evaporation process of the droplets in a stagnant gas space. They discussed the process based on the molecule diffusion on the spherical droplet surface in constant temperature.

Apart from evaporation, crystallization is another important process of the ZLD system. The crystallization period could affect the size and the porosity of the crystals directly. From Ford's work [13], the high crystallization rate could lead to high porosity and larger size of the crystals when evaporating a spherical droplet. Furthermore, different parameters [14], like the size, the porosity and the compounds of the particle, lead to different electric penetration for the electrostatic precipitator in the coal-fired power plant. Thus, it is significant to study the crystallization process of the desulfurization wastewater due to the efficiency of the collection by the electrostatic precipitator in Ran and Zhang's design [9].

For the porosity and the morphology of the crystals from the evaporated droplet, the Peclet (Pe) number [15–17] was used to analyze them. With the increase of the Pe number, the crystals [15] from evaporation spherical droplet could be more likely to form thick shell and smaller holes on the surface, because the mobility [16] of the particle was larger. Also, in Baldwin et al's work [17], the Pe number affected the deposition of the sessile droplet. Lower Pe number gave more uniform deposition which means lower value of the porosity and small size of crystals.

For the crystallization process, some achievements were also reported. Landfester et al. [18,19] detected the change in crystallization temperature with different concentration of solutes in a single miniemulsion droplet using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) methods. Liang et al. [20] studied the evaporation and crystallization characteristics of the desulfurization wastewater and investigated the relationship between the crystallization rate and the porosity of the crystals, at different operating conditions with the TGA-DSC method. Furthermore, in the gas flue at the end part of the boiler, there are lots of fly ash [21] in it. This fly ash could interact with the droplet and affected the evaporation and crystallization