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Systematic approach to determination of optimum gas-phase mass transfer rate for high-gravity carbonation process of steelmaking slags in a rotating packed bed

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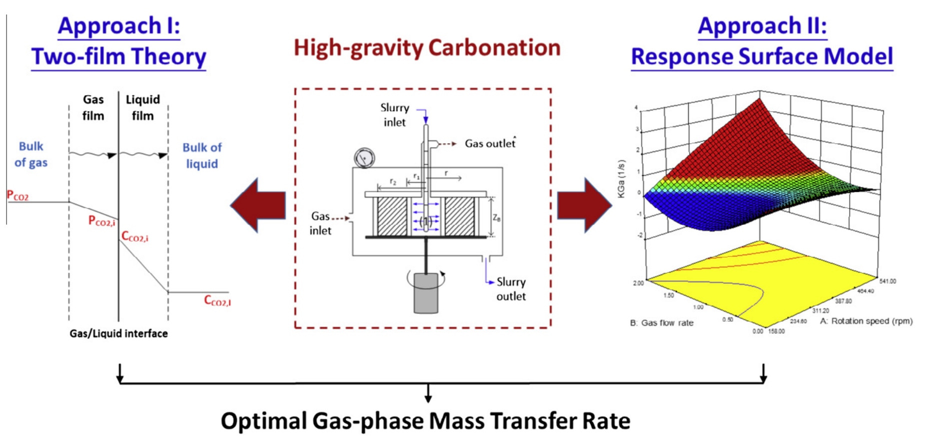
HIGHLIGHTS

- Mass transfer models of carbonation in RPB were developed based on two-film theory.
- Correlation between dimensionless groups and K_Ga/HTU was experimentally determined.
- Effects of different operating variables such as ω, Q_s, Q_G, and L/S were evaluated.
- RSM was introduced for confirming mass transfer model from statistic point of view.
- Optimal operating modulus were graphically determined based on both models and RSM.

GRAPHICAL ABSTRACT



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ABSTRACT

In order to reduce CO_2 emissions and waste generation from the steelmaking industry, a high-gravity carbonation process via rotating packed bed (RPB) was developed using cold-rolling mill wastewater (CRW) and basic oxygen furnace slag (BOFS). Since mass transfer among phases is believed to be a key to effective carbonation for CO_2 fixation, in this study, a mass transfer model for the high-gravity carbonation process was developed based on two-film theory. The mass transfer characteristics including overall gas-phase mass transfer coefficient (K_Ga) and height of a transfer unit (HTU) were determined accordingly. The results indicated that the mass transfer resistance of carbonation using BOFS/CRW in an RPB was mainly lay on the liquid side. In addition, the effect of key operating variables such as rotating speed, slurry flow rate, gas flow rate, and liquid-to-solid (L/S) ratio on mass transfer characteristics was evaluated. The developed model was validated with the experimental data, where the experimental K_Ga values lay within ±20% of the values estimated. Based on the obtained results, empirical models of K_Ga and HTU values were established. Furthermore, response surface methodology (RSM) was applied to optimize the high-gravity carbonation process from the viewpoint of mass transfer characteristics. The obtained

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RSM results were in fairly good agreement with the results of the developed model based on the two-film theory. Based on the theoretical models and statistical analyses, the optimum gas-phase mass transfer rate for high-gravity carbonation process of steelmaking slags in an RPB was graphically determined.

1. Introduction

The CO_2 mineralization by accelerated carbonation has been regarded as diffusion controlled reaction (i.e., mass-transfer limited) according to the findings reported in the literature [1–3]. Therefore, different types of approaches such as physical intensification [4] and bio- or chemical activation [5,6] were recently carried out to improve the mass transfer and reaction kinetics. Based on this idea, a rotating packed bed (RPB) reactor has been introduced to improve the mass transfer rate among phases due to its high centrifugal forces and great micro-mixing ability. RPB, the so-called "high-gravity" (sometimes called the "HIGEE") process, is designed to generate high acceleration via centrifugal force, thereby enhancing mass transfer between gas and liquid, and even between liquid and solid. Since RPB can provide a mean acceleration of hundreds, and even thousands, of times greater than the force of gravity, it can effectively lead to the formation of thin liquid films and micro- or nano-droplets [7–9]. Therefore, the volumetric gas–liquid mass transfer coefficients are an order of magnitude higher than those in a conventional packed bed, leading to dramatic reductions in equipment size over that required for equivalent mass-transfer in a gravity flow packed bed [7,9,10].

According to previous studies [11–13], performance of the high-gravity carbonation process using an RPB was found to be better than that using an autoclave or slurry reactor. The CO_2 removal efficiency in the flue gas by alkaline steelmaking wastes in the RPB process was more than 96% with a short retention time (less than 1 min) under ambient temperature and pressure conditions [13]. Precipitation is usually a very fast process and the mixing, especially micro-mixing, of the process is very important for particle size distribution [14]. In such a case, the precipitation process would be controlled by the intrinsic reaction kinetics due to both the excellent micro-mixing [15] and the fine BOFS with a particle size of 44–88 μm in an RPB [11,13,16]. Therefore, before the passive layers around particles are formed, the minerals in BOFS can be rapidly dissolved into solution when the BOFS moved though the packed bed. In other words, the mass transfer between liquid and solid phases may not be the rate limiting factor in the high-gravity carbonation system. In addition, the reaction mechanisms and kinetics of carbonation in an RPB have been studied using shrinking core model [11] and surface coverage model [17], which indicate that the film diffusion should be one of the key rate-limiting steps in the high-gravity carbonation system.

Over the past years, several theoretical models have been developed for describing mass transfer phenomena of RPB for various applications, especially for gas–liquid separation [18,19], which was the earliest designed process. However, sophisticated assumptions and complicated partial differential equation programming were generally required. In addition, no major studies on gas–liquid or liquid–solid mass transfer for the high-gravity carbonation process of alkaline solid wastes have appeared in the literature so far. Therefore, development of an accurate and precise model in which only algebraic equations are included is essential to optimize the high-gravity carbonation process.

The objectives of this study were to (1) develop mass transfer models for high-gravity carbonation of alkaline wastes in an RPB based on theoretical theory, (2) determine the mass transfer characteristics such as the overall gas-phase mass transfer coefficient (K_Ga) and height of a transfer unit (HTU); (3) evaluate the effect of key operating variables such as rotating speed, slurry flow rate and liquid-to-solid (L/S) ratio on mass transfer characteristics; (4) establish a statistical prediction model via response surface methodology (RSM) using experimental data, and (5) optimize the high-gravity carbonation process through a graphical presentation according to the results of the theoretical models and RSM.

2. Materials and methods

2.1. Materials

The ground basic oxygen furnace slag (BOFS) was provided by China Steel Corporation (CSC) (Kaohsiung, Taiwan). The ground BOFS was dried in an oven at 105 °C overnight to eliminate moisture and then stored at room temperature in large airtight containers. The specific gravity of BOFS was 3.14 g/cm^3, with a mean particle size of 12.7 μm. The BOFS was rich in CaO (~43%) and Fe_2O_3 (~29%), with minor component of SiO_2 (~13%) and MgO (~6%). The $Ca(OH)_2$ and free-CaO contents were found to be 7.7% and 1.0%, respectively.

Cold-rolling mill wastewater (CRW) was introduced in the carbonation reaction. The pH value of the CRW was in the range of 8.9–12.2. The major chemical components of CRW were sodium ions (~860 mg/L), potassium ions (~93 mg/L), chloride ions (~2430 mg/L) and sulfate ions (~240 mg/L). Moreover, the concentrations of calcium and magnesium ions were around 4.5 ppm and 0.5 ppm, respectively, of which their contribution to CO_2 mineralization could be neglected. The conductivity of the fresh CRW was approximately 10,000 μmho/cm. It was noted that the CRW can increase the leaching rate and capacity of calcium ions from BOFS into solution, thereby enhancing the carbonation efficiency [12,16,20].

2.2. Specifications of rotating packed bed

In this study, high-gravity *ex-situ* carbonation was performed in a small pilot-scale RPB. The rotation type of packed bed in this study was horizontal rotation, where the gas and slurry were mixed counter-currently within the RPB. The packing zone, where stainless steel wire acted as a packing material, had an inner diameter of 20.5 cm and an outer diameter of 62.5 cm, corresponding to an arithmetic mean radius of 46.5 cm. The axial height of the packed bed was 19.9 cm. In this case, the designed capacity of gas flow in the RPB was around 2.5 m^3/min. In addition, the liquid distributor consisted of a tube with two vertical sets of holes spaced 180° apart. Each hole had a diameter of 0.1 cm, and the holes were spaced at intervals of 0.6 cm. Moreover, the designed liquid loading was 0.009–0.021 m^3/(m^2 min).

2.3. Carbonation experiments

In the experiments, *ex-situ* carbonation was carried out by the high-gravity RPB process using a hot-stove gas at the No. 3 blast furnace plant in CSC (Kaohsiung, Taiwan). The flue gas of the hot