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Performance evaluation of integrated air pollution control with alkaline waste valorization via high-gravity technology

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ABSTRACT

In this study, a high-gravity process using alkaline wastes, i.e., petroleum coke fly ash, was proposed for integrated air pollution control, including nitrogen oxides, carbon dioxide and particulate matters. After the control process, the reacted fly ash can be further utilized as supplementary cementitious materials in cement mortar to realize a circular economy. The performance of air pollution control using petroleum coke fly ash in a high-gravity process was evaluated by on-site operations. The results indicated that the removal ratios of nitrogen oxides, carbon dioxide and particulate matters in the flue gas were approximately 99% (in the presence of ozone), 95% and 80%, respectively. In addition, the kinetics of carbon dioxide removal in the high-gravity process was analyzed by a surface coverage model. A high reaction rate constant of 0.27 mol/min/m^2 was reached under a liquid-to-solid of 20 mL/g and a rotating speed of 1000 rpm at 20 °C. Furthermore, the performance of reacted fly ash used in blended cement mortar was evaluated. Compared with the pure Portland cement mortar, the compressive strength of cement mortar with reacted fly ash can be increased by 26%. Lastly, a preliminary economic analysis was conducted to estimate the costs and benefits from the process.

1. Introduction

Conventional air pollutants, including nitrogen oxides (NO_x) and particulate matter (PM), are still leading to massive concerns on environmental deterioration. The commonly used technology for NO_x removal in industries is selective catalytic reduction. Although the NO_x removal efficiency by the selective catalytic reduction could typically reach 90%, it requires a great amount of energy for reactor heating to maintain appropriate operations and longevity of catalysts [1]. Available PM removal technology includes venturi scrubber, cyclone, baghouse filter and electrostatic precipitator. The mechanisms of PM removal are either through diverging particles from the trajectory of the gas stream (e.g. cyclone and electrostatic precipitator), or by filtration (e.g., wet scrubber and baghouse filter) [2]. These PM removal technologies normally lead to a considerable pressure drop and high maintenance costs. In other words, it is necessary to develop an integrated air pollution control process that could simultaneously remove several kinds of air pollutant emissions in the flue gas, even for the greenhouse gases such as carbon dioxide (CO_2).

Industries are seeking for pathways by which wastes can be recovered and reused. For instance, petroleum coke fly ash (PCFA) has been widely used as supplementary cementitious materials (SCM) since it exhibits great pozzolanic properties that can facilitate hydration reactions for cement mortar [3]. However, the content of lime (free-CaO) in fresh PCFA might cause potential concerns and impacts on durability of construction materials. Since the active alkaline compounds such as lime and magnesia could be neutralized and stabilized by acidic pollutants (e.g., SO_x and CO_2) from the flue gas [4], PCFA can be potentially used as the feedstock for integrated air pollution control. For instance, the CO_2 in the flue gas would react with lime in PCFA via accelerated carbonation reaction to form calcium carbonate precipitates [5]. Accelerated carbonation is also regarded as a treatment process for the immobilization of toxic elements, such as Cr and Pb, from alkaline solid wastes such as steel slags and fly ash [6,7]. As a result, a great number of benefits can be realized if PCFA is used for integrated air pollution control: (i) simultaneous removal of

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various air pollutants such as NO_x and PM; (ii) reduction of CO_2 emission in the flue gas; (iii) improvement of physico-chemical properties of PCFA for further utilization.

To improve the mass transfer between the gas and liquid phases, a high-gravity (HiGee) process provides a holistic solution that integrates air pollution control and waste valorization. The HiGee process generates a high gravitational field through high-speed rotation, enhancing mass transfer between aqueous and gaseous phases [8,9]. Tiny droplets are generated within the HiGee process when subject to intensified gravitational field, thereby increasing the number of collision between droplets and PM [10,11]. The HiGee technology has been successfully combined with accelerated carbonation to enhance the mass transfer between CO_2 and liquid phase, known as a high-gravity carbonation process [12,13]. Compared with conventional reactors such as an autoclave or a slurry reactor, the high-gravity carbonation process can significantly increase the efficiency and capacity of CO_2 mineralization using alkaline wastes [14,15]. The reacted alkaline wastes also can be valorized as high value-added products such as green construction materials [16]. Under this framework, a waste-to-resource supply chain can be established for achieving a circular economy system.

To the best of our knowledge, this article should be considered as a pioneering study combining integrated air pollution control including PM, NO_x, and CO_2 with industrial alkaline waste stabilization using PCFA in the HiGee process. The objectives of this study were to (1) evaluate the performance of the HiGee process for integrated air pollution control including PM, NO_x, and CO_2, (2) determine the kinetics of accelerated carbonation using PCFA for CO_2 mineralization by a surface coverage model, (3) examine the feasibility of using carbonated PCFA as SCM in cement mortar, and (4) conduct a preliminary economic analysis for integrated air pollution control and alkaline waste valorization using the HiGee process.

2. Materials and methods

2.1. Integrated air pollution control using PCFA via HiGee

In this study, a HiGee process was applied for integrated air pollution control using PCFA at a petrochemical plant in Taiwan. The HiGee process was equipped with a counter-current rotating packed bed reactor with a packing size of 25.4 cm in height and 28.0 cm in width. In order to enhance the resistance against the acid (SO_x and NO_x) corrosions, the HiGee is made up of SS-316 stainless steel. The PCFA was mixed with blowdown wastewater (BDW) in the slurry tank at the liquid-to-solid (L/S) ratio of 10 m^3/kg. Then, the PCFA slurry was pumped into the rotating packed bed at a constant flow rate of 0.02 m^3/min, together with the flue gas driven by a blower in a countercurrent direction. The pH of PCFA slurry was measured and recorded by a pH meter (InPro 4260, Mettler Toledo, Switzerland) since the removal of NO_x and CO_2 is highly related to the pH value of the slurry. For the CO_2 removal in the flue gas, the reaction would complete once the pH of slurry drops below 6.3, according to the equilibrium constant of $HCO_3^-/H_2CO_3^*$ system [17]. The reacted PCFA slurry was discharged into the sedimentation tank, and then the separated PCFA was collected for utilization as supplementary cementitious material.

2.2. Characterization of PM emissions in flue gas

PM is considered as one of the critical air pollutants in flue gas, and could be collected by the HiGee process. In this study, PM in the flue gas was collected by a micro-orifice uniform deposit impactor (MOUDI, MSP, US) and weighed by an electronic balance (AUW220D, Shimadzu, Japan). In this way, weight fraction of particles with different particle sizes could be obtained. Before and after weighing, the impactors were conditioned under 120 °C for 24 h to reduce the disturbance from volatile substances.

In general, particle size distribution follows the Gaussian distribution regime, which may bring arbitrariness in the size distribution analysis. For this reason, a probability-log plot is used, where the relationship of cumulative distribution versus particle size is presented by a straight line [18]. Subsequently, the parameter of cut diameter, i.e., the diameter corresponding to 50% of cumulative curve, can be directly determined from the plot. In addition, geometric standard deviation (σ_{gm}) can be calculated to evaluate the variation extent of size distribution:

$$\sigma_{gm} = \frac{50\%\ \text{size}}{85\%\ \text{size}} \tag{1}$$

where σ_{gm} is the geometric standard deviation; 50% size and 85% size are 50% (cut diameter) and 85% weight fraction determined from the plot, respectively.

According to the difference in weight of particles at the inlet and outlet of the HiGee process, the fractional removal efficiency of PM for different particle sizes can be obtained. By multiplying fractional efficiency and the corresponding weight fraction, the overall PM removal efficiency can be calculated by Eq. (2):

$$E_{overall} = \sum_{i=1}^{12} E_i \times f_i \tag{2}$$

where $E_{overall}$ is the overall removal efficiency, E_i and f_i are fractional removal efficiency and weight fraction of the particle with aerodynamic diameter i, respectively.

2.3. Evaluation of removal efficiencies of NO_x and CO_2 in flue gas

A gaseous composition analyzer (PG-350, HORIBA, Japan) was used for monitoring the concentrations of NO_x, CO_2 and O_2 in flue gas. The resolutions of O_2 and CO_2 were 0.01 vol.%, while 0.01 ppm for both SO_2 and NO/NO_x. The removal efficiency of NO_x and CO_2 in the flue gas by the HiGee process can be determined by Eq. (3):

$$\eta\ (\%) = \frac{Q_i \times \rho \times C_i - Q_o \times \rho \times C_0}{Q_i \times \rho_i \times C_i} \times 100\% \tag{3}$$

where η (%) is the removal efficiency of a specific pollutant; Q_i (m^3/min) and Q_o (m^3/min) are the flow rate of inflow and outflow gas, respectively; and ρ (kg/m^3) is the density of flue gas.

Based on the removal efficiency, the daily load (i.e., removal capacity) of a certain pollutant removal can be estimated by Eq. (4):

$$\text{Daily load} = \frac{Q \times \eta \times M_{CO_2} \times T_m \times P_{SC}}{22.4 \times T_{SC} \times P_m} \times 1440 \tag{4}$$

where T (°C) is the temperature, and P (Pa) is the pressure of flue gas. The subscripts of m and sc indicate the measured and the standard values, respectively.

To quantify the amount of CO_2 fixed in PCFA, the weight loss due to thermal decomposition of $CaCO_3$ products in carbonated PCFA was determined by a thermo-gravitational analysis (TGA, STA 6000, PerkinElmer, US). In TGA, temperature range was set from 50 to 950 °C with a heating rate of 10 °C/min under a nitrogen atmosphere.

2.4. Determination of CO_2 mineralization kinetics

To determine the reaction kinetics, the PCFA slurry with different L/S ratios, i.e., 10, 15, 20, and 25 mL/g, was stirred constantly at 650 rpm and pumped to the inner edge of the HiGee reactor at

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