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Effects of water vapor pretreatment time and reaction temperature on CO_2 capture characteristics of a sodium-based solid sorbent in a bubbling fluidized-bed reactor

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Abstract

CO_2 capture from flue gas using a sodium-based solid sorbent was investigated in a bubbling fluidized-bed reactor. Carbonation and regeneration temperature on CO_2 removal was determined. The extent of the chemical reactivity after carbonation or regeneration was characterized via ^{13}C NMR. In addition, the physical properties of the sorbent such as pore size, pore volume, and surface area after carbonation or regeneration were measured by gas adsorption method (BET). With water vapor pretreatment, near complete CO_2 removal was initially achieved and maintained for about 1–2 min at 50 °C with 2 s gas residence time, while without proper water vapor pretreatment CO_2 removal abruptly decreased from the beginning. Carbonation was effective at the lower temperature over the 50–70 °C temperature range, while regeneration more effective at the higher temperature over the 135–300 °C temperature range. To maintain the initial 90% CO_2 removal, it would be necessary to keep the regeneration temperature higher than about 135 °C. The results obtained in this study can be used as basic data for designing and operating a large scale CO_2 capture process with two fluidized-bed reactors.

Keywords: CO_2; Capture; Dry sorbent; Carbonation; Fluidized-bed reactor

1. Introduction

The concentration of CO_2 in the earth's atmosphere is increased by combusting fossil fuels to generate electricity. Recently, research in CO_2 capture and storage has attracted increasing attention across a variety of industrial fields (Metz et al., 2005). Several methods have been suggested for CO_2 capture. These include wet absorption, adsorption, membrane separation, and cryogenic distillation. However, these methods need to overcome the limits of cost and energy required to treat the large flue gas streams from fossil fuel-fired power plants. Recently CO_2 capture using dry regenerable sorbents has been studied as an innovative concept for CO_2 recovery (Fauth et al., 2004; Liang et al., 2004; Abanades et al., 2005; Ryu et al., 2005). Nelson et al. (2005) presented some results from a circulating fluidized-bed reactor. Yi et al. (2005, 2007) and Yi et al. (2006) have shown CO_2 reaction characteristics of dry sorbent in a bubbling fluidized-bed reactor or in the continuous solid circulating mode between a fast fluidized-bed carbonator and a bubbling fluidized-bed regenerator. This study focused on the performance of solid sorbent in a bubbling fluidized-bed reactor.

CO_2 is efficiently removed from the flue gas stream by reaction with solid sorbent while regeneration produces an off-gas containing CO_2 and H_2O for sodium- or potassium-based sorbent or only CO_2 for calcium-based sorbent. The condensation of an off-gas generates highly pure CO_2, which is suitable for chemical feed stock or storage. Because solid sorbents are derived from alkali metals and carbonated

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solids can be regenerated solely from the heat in the flue gas stream, the solid sorbent process for CO_2 capture is expected to be cost-effective and energy-efficient (Liang et al., 2004). In the solid sorbent process, heat control is important to avoid hot spots generated during the highly exothermic carbonation reaction, and high superficial velocity is necessary to reduce reactor size. To meet these requirements a fluidized-bed reactor could be one of optimum candidates for the CO_2 capture process using dry sorbents. The fluidized-bed reactor can provide high heat and mass transfer rates between gas and particles, easily remove heat produced during exothermic reaction, and maintain isothermal conditions through the reactor due to rapid mixing. Thus the fluidized-bed reactor system would be suitable for large-scale operations (Kunii and Levenspiel, 1991).

The present work investigated the CO_2 capture characteristics and performance of a dry sorbent in a bubbling fluidized-bed reactor in order to obtain the basic operational parameters, which will be valuable for designing and operating a large scale CO_2 capture process with two fluidized-bed reactors. The effect of carbonation temperature, water vapor pretreatment and regeneration temperature on CO_2 removal was studied to check the capability of the sorbent. The extent of conversion of active component in solid sorbent particles after carbonation or regeneration was confirmed via ^{13}C NMR (nuclear magnetic resonance). In addition, the physical properties of the sorbent such as pore size, pore volume, and surface area after carbonation and regeneration were measured by BET.

2. Experimental

2.1. Materials and apparatus

Fig. 1 shows the schematic diagram of the experimental apparatus for carbonation and regeneration, including a bubbling fluidized-bed reactor. The apparatus consists of a gas injection part, reactor, bubbler for water vapor generation, gas post-treatment part, and gas analyzer. A reactor with an inner diameter of 0.05 m and a height of 0.8 m was made of quartz and placed inside of a furnace. Reactor temperature was controlled by a furnace and a temperature controller and measured by thermocouples fitted in the reactor. To prevent an abrupt rise in temperature and to keep the temperature constant, cold air was blown to the reactor during the carbonation reaction. Each gas flow was quantitatively controlled with a mass flow controller (Brooks, Japan). Effluent gas from reactor firstly passed through a condenser to remove H_2O, then passed through a filter to remove dust, and finally, to a gas analyzer (ABB, USA) which analyzes CO_2 every 10 s.

The spray-dried solid sorbent, Sorb NX35, used in this study was provided by KEPRI (Korea Electric Power Research Institute). It consists of 35% Na_2CO_3 as an active ingredient and 65% support and binder for mechanical strength as well as pore structure. More details of sorbent preparation can be found in the other literature (Ryu et al., 2005). Sorb NX35 has a bulk density of 750 kg m^{-3}, a mean particle diameter of 1.046×10^{-4} m, and an attrition index of 1.0, as defined by the ASTM (American Standard Testing Material) D5757-95 procedure, which corre-sponds to the 5-h loss.

2.2. Procedure

Taking the real operation conditions in a large scale fluidized-bed reactor into account, 0.09 kg of the sorbent was placed into the reactor to maintain a residence time of gas mixture of 2 s and a superficial velocity of 0.03 m s^{-1}. To simulate real flue gas composition, a gas mixture of 10% CO_2, 77.8% N_2 and 12.2% H_2O was used. The water vapor needed for carbonation was fed by passing the mixture gas

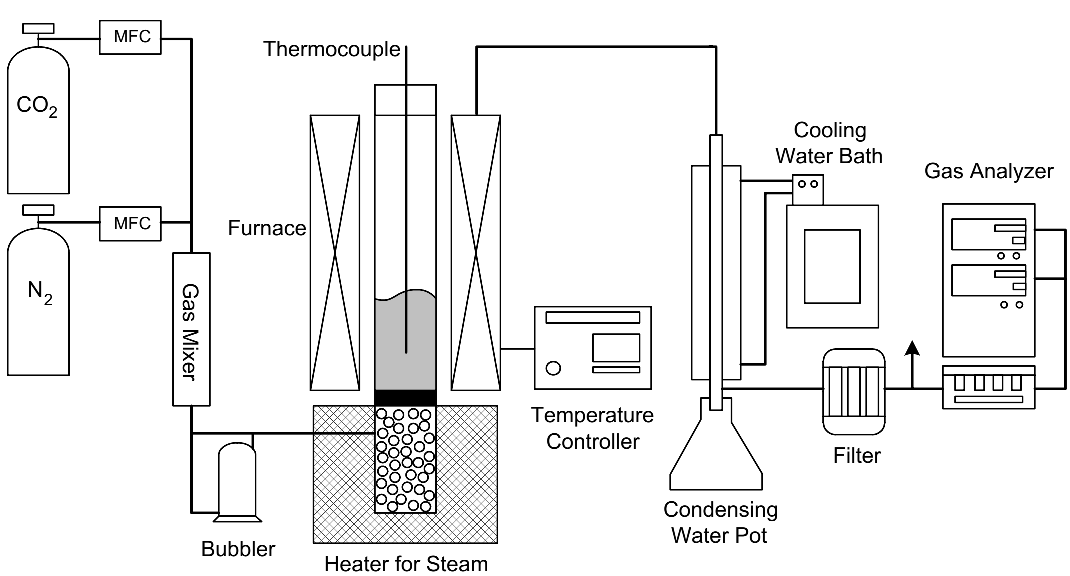

Fig. 1. The schematic diagram of experimental apparatus for carbonation and regeneration.