Demonstration of Carbon Dioxide Capture from Coal-fired Flue Gas Using Calcium-based Sorbents

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Abstract

Carbon dioxide (CO_2) is one of the most important greenhouse gases causing global climate change. To minimize the undesirable impact of carbon dioxide emissions on the environment, many new technologies are being developed to reduce its emissions from the combustion of fossil fuels. A promising technology is the multi-cyclic Carbonation and Calcination Reaction (CCR) process. The CCR process utilizes a calcium-based sorbent in a high temperature reaction (carbonation) to capture the CO_2 from the flue gas stream. The subsequent calcination reaction releases a pure, dry stream of CO_2, which can be sequestered. A 20 pph-scale facility firing coal has been constructed at The Ohio State University to demonstrate the CCR process. A number of calcium-based sorbents were tested at this facility. The effect of residence time and Ca/C ratio were evaluated and over 90% CO_2 capture was achieved on a once-through basis. One of the calcium based sorbents has exhibited stable reactivity for CO_2 capture over multiple cycles.

Introduction

CO_2 is one of the major greenhouse gases contributing to global climate change. In the United States, coal-combustion for electricity generation is responsible for approximately one-third of anthropogenic carbon dioxide emissions.

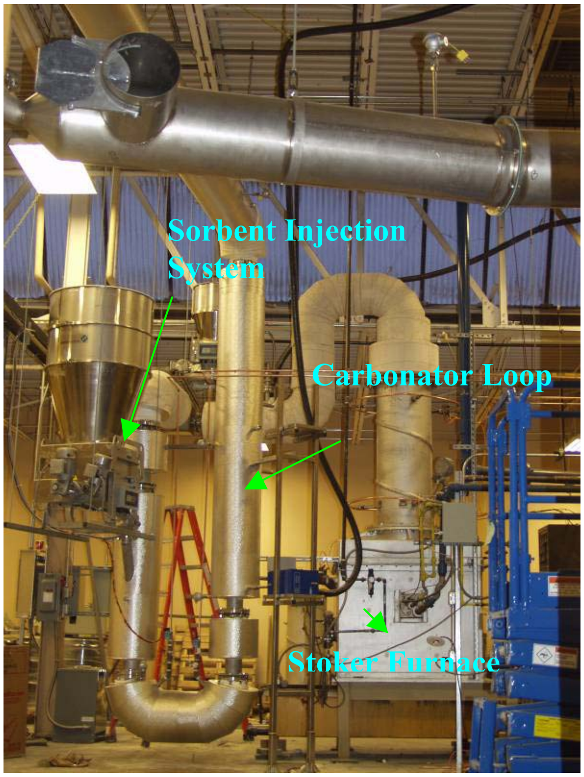
There are three main approaches to capturing CO_2 generated from fossil fuels: pre-combustion capture system, post-combustion capture system and oxy-combustion [1]. Pre-combustion capture systems process the primary fuel in a reactor to produce separate streams of CO_2 for storage and H_2, which is used as a fuel. Post-combustion system separates CO_2 from flue gas generated by combustion of fossil fuels. Oxy-combustion uses oxygen instead of air for combustion, producing a flue gas that is mainly H2O and CO_2, which is readily captured. Both pre-combustion and oxy-combustion produce a CO_2 stream by making significant design changes to a conventional boiler, which is a good strategy for new power plants. For existing electric power plants, post-combustion capture might be the best option.

A number of technologies are currently being developed to separate CO_2 from coal combustion flue gas. These include sorbents/solvents separation, membrane separation and cryogenic distillation. The calcium based solid sorbents technology has received increasing attention [2-5]. This technology utilizes a calcium-based sorbent in a carbonation reaction at high temperature to capture the CO_2 from the flue gas stream and releases a pure, dry stream of CO_2 in the subsequent calcination reaction for sequestration. One of the advantages of this technology over low temperature process (such as amine scrubbing) is that the heat released at the temperatures of 600~700 C during the carbonation process is utilized.

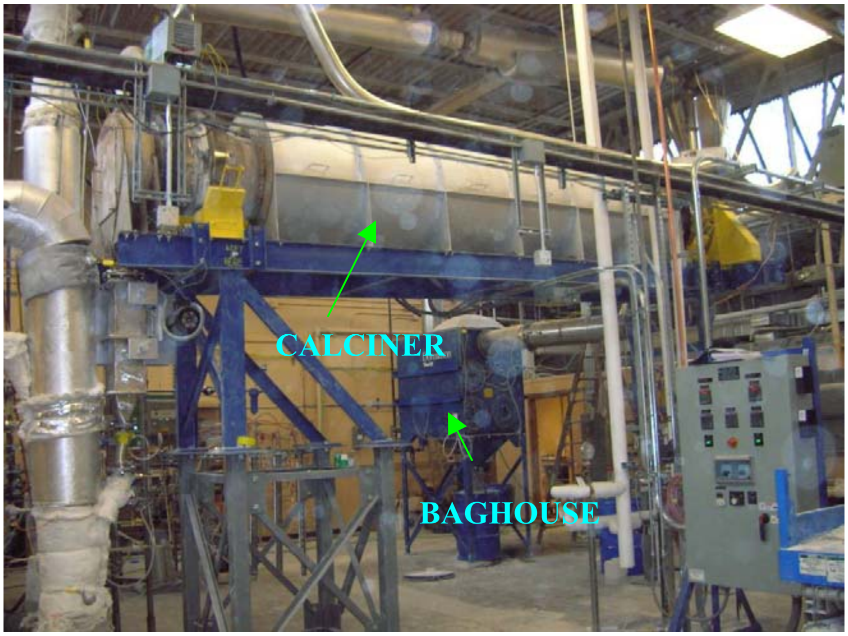
In this work, a 20 pph-pilot scale facility was constructed to demonstrate the calcium-based carbonation/calcination reactions (CCR) process. The objective of this work includes (1) testing the feasibility of this process; (2) determining CO2 capture capacity of different calcium-based sorbents over multi-cycles; and (3) determining the effect of residence time and calcium/carbon ratios on CO_2 removal.

Test facility

The experiments were conducted in the facility shown in Figure 1. The system consists of a 20lb/hr coal-fired stoker furnace, a rotary calciner, sorbent and ash injection systems, upstream and downstream gas sampling systems, fans, particulate control devices and data acquisition, control systems and associated instrumentation. The sorbent is injected at temperatures between 600-700 C, which is the optimal temperature window for the carbonation reaction. The carbonated sorbent is then separated from flue gas and sent to the rotary calciner for regeneration. The calciner regenerates the spent sorbent for a subsequent CO_2 capture cycle. The calciner, which is 20 ft. long and 2 ft in diameter, is electrically-heated. The maximum calciner operating temperature is 980 C.



(a)



(b)

Figure 1 Demonstration facility for CO_2 capture

Test results

Figure 2 shows the effect of increasing Ca/C ratio on CO_2 removal on a once-through basis. It can be seen that CO_2 removal increases with an increase in Ca/C ratio. Among the three tested calcium based sorbents, sorbent 1 gave the highest CO_2 capture. CO_2 removal efficiency significantly increases from 46% to nearly 100% as Ca/C ratio increases from 0.6 to 1.6. However, CO_2 removals by sorbents 2 and 3 seem less sensitive to the variation of Ca/C ratio in comparison to sorbent 1.

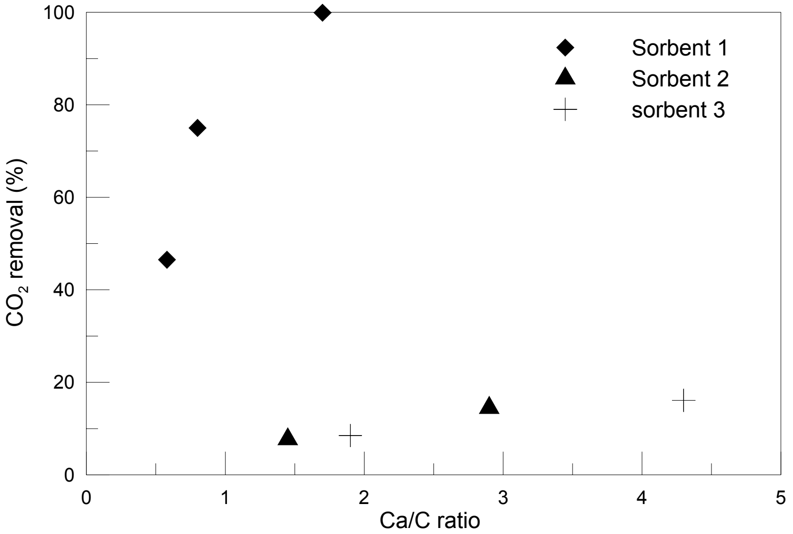


Figure 2. Effect of Ca/C ratio on CO_2 removal

Figure 3 presents the effect of the residence time on CO_2 removal on a once-through basis. It is obvious that the residence time has a significant impact on the CO_2 removal. CO_2 removal increases from 38% to 55% as the residence time is doubled.

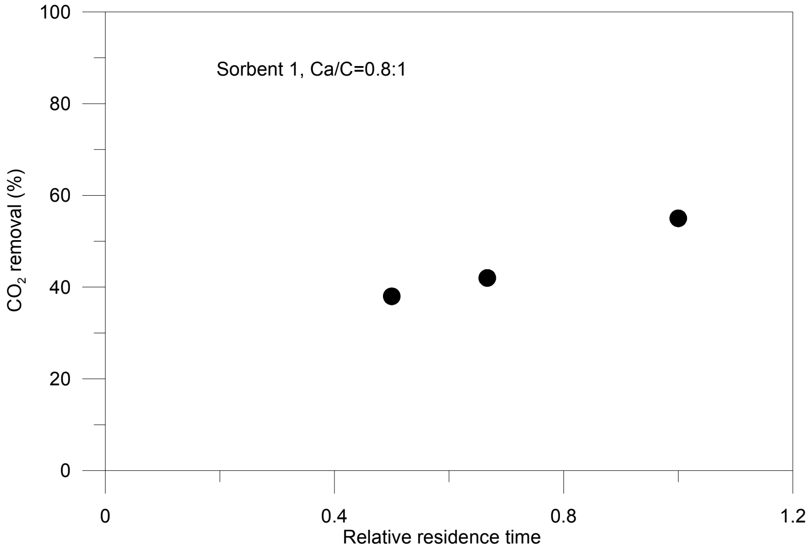


Figure 3 Effect of residence time on CO_2 removal

A critical issue for this process is the stability of sorbent reactivity after multiple cycles. No obvious decay was observed for sorbent 1 after several cycles. We are continuing to conduct recycle tests to confirm sorbent 1 recycle performance.

Summary

A 20lb/hr combustion facility was constructed at The Ohio State University for the purpose of demonstrating the CCR process. Three types of calcium based sorbents were tested on a once-through basis. Sorbent 1 achieved greater than 90% CO_2 capture at Ca/C ratio of 1.6. Multi-cyclic CO_2 capture test show that there is no obvious decay of sorbent 1 after three recycles.

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References

[1] Working group III of the Intergovernmental panel on climate change (2005), “IPCC special report on carbon dioxide capture and storage”.

[2] Iyer, Mahesh V.; Gupta, Himanshu; Sakadjian, Bartev B.; Fan, Liang-Shih. (2004), “Multicyclic Study on the Simultaneous Carbonation and Sulfationof High-Reactivity CaO”*Ind. Eng. Chem. Res.*, pp.: 3939-3947.

[3] Sun, Ping; Grace, John R.; Lim, C. Jim; and Anthony, Edward J. (2007), “Removal of CO_2 by Calcium-Based Sorbents in the Presence of SO_2”, *Energy & Fuels*. pp:

[4] Alvarez, Diego and Abanades, J. Carlos. (2005), “Pore-Size and Shape Efects on the Recarbonation Performance of Calcium Oxide Submitted to Repeated Calcination/Recarbonation Cycles”, *Energy & Fuels.* pp:.

[5] Gupta, Himanshu and Fan, Liang-Shih. (2002), “Carbonation-Calcination Cycle Using High Reactivity Calcium Oxide for Carbon Dioxide Separation from Flue Gas” *Ind. Eng Chem Res*. pp: