A Low Cost, High Capacity Regenerable Sorbent for CO_2 Capture

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INTRODUCTION

Coal currently accounts for nearly 56% of U.S. electric power generation, and since the U.S. has 25% of world's coal reserves,[1] and will play an increasingly important role in meeting the Nation's future energy needs. Coal contains a variety of elements in addition to carbon and hydrogen, and as a result coal based power plants contribute approximately 2/3rd of the country's sulfur dioxide (SO_2) emissions, 1/5th of the nitrogen oxides (NO_x) emissions, over 1/3rd of the mercury emissions and 36% of U.S. carbon dioxide (CO_2) emissions. CO_2 is a major greenhouse gas and plays a significant role in global climate change. Concerns about global climate change caused by greenhouse gas emissions could lead to future regulations on the CO_2 emitted by coal-fired power plants.

Advanced power generation cycles, such as the Integrated Gasification Combined Cycle (IGCC), are likely to replace conventional pulverized-coal combustion plants in new construction due to their higher efficiency and lower emissions. IGCCs use a gasifier to convert coal into fuel gas and use a combined cycle power generation block to generate electricity. With a projected 50% increase (from 300 GW to 450 GW) in electricity demand by 2030,[2] the vast bulk of the electricity produced both in the U.S. and world-wide will continue to come from pulverized coal (PC). Therefore, if in the future CO_2 emissions are regulated existing PC coal power plants will need to be retrofitted with a low cost CO_2 capture technology that can efficiently remove CO_2 from the dilute low-pressure flue gas stream and in the case of IGCC it makes sense to capture it early in the IGCC process (before the turbine) where it is concentrated and at high pressure. Since it is far easier and less expensive to remove impurities when they are at high pressure and concentrated in the hot coal gas than when they are at atmospheric pressure and have been diluted by more than 10:1 in the combustion turbine. This is true not only of minor components such as sulfur, nitrogen and metals, but of carbon (CO_2) as well.

TDA's CO_2 SORBENT

TDA is developing a novel sorbent that removes CO_2 via physical adsorption from both synthesis gas and flue gas, while the relatively strong affinity of the sorbent to CO_2 enables effective operation at temperatures up to 300^oC (well above the dew point of synthesis gas stream generated by commercial gasifiers). However, because the sorbent and the CO_2 do not form a true covalent bond, the energy needed to regenerate our sorbent (5.4 kcal per mol of CO_2) is much lower than that observed for either chemical absorbents (e.g., 29.9 kcal/mol CO_2 for sodium carbonate) or amine-based solvents (e.g., 14.2 kcal/mol CO_2 for monoethanolamine). Our sorbent can be regenerated isothermally and CO_2 could be recovered at pressure (~150 psia).

Thus, the energy needed to regenerate the sorbent and compress the CO_2 for sequestration is significantly lower than that for any technology reported to date. TDA's CO_2 sorbent could be used to capture CO_2 in both Integrated Gasification Combined Cycle (IGCC) and pulverized coal (PC) fired power plants. The high surface area and favorable porosity of the sorbent also provides a unique platform to introduce additional functionality, such as active groups to catalyze the water-gas-shift (WGS) reaction or to remove trace metals (e.g., Hg, As).

Experimental Details

We carried out initial adsorption experiments by using a Shimadzu TGA-50 Thermogravimetric Analyzer, where the sorbent is placed in a microbalance and weight change of the sample due to its exposure to a gas containing CO_2 is correlated to the CO_2 adsorption capacity. In these tests, a small quantity of sorbent was placed into the sample holder where it is possible to control the temperature, flow rate and composition of the gases. We used gas streams containing pure CO_2 and CO_2/N_2 mixtures (the operating pressure of the analyzer was limited to 15 psig; the use of pure CO_2 allowed us to simulate higher pressure) to simulate the adsorption step. The CO_2 adsorption/desorption capacity was measured from the weight change of the sample. For desorption we switched the gas flow to pure N_2. Although in the actual system a steam sweep will be used to generate the CO_2 partial pressure difference, we could not feed steam to the existing TGA setup. Assuming the steam will be as inert as nitrogen, we used TGA to provide a quick insight on the overall CO_2 capacity and rate of absorption and desorption for the sorbent. In these initial tests, we varied the operating temperature to study the effect of temperature and also to measure the heat of adsorption.

Results and Discussion

In the TGA tests, we varied the temperature of the sorbent (at P_{CO2}= 9.7 psia) to evaluate the impact of temperature on capacity (Figure 1). As expected the sorbent achieved a much higher capacity at 120^{o}C (1.65% wt.) than at 220^{o}C (0.36% wt.). It is important to realize that the CO_2 partial pressure in these tests was at 9.7 psia or 0.66 atm), an order of magnitude lower than in gasifier streams (our TGA cannot be pressurized). Much higher CO_2 capacities can be achieved at higher pressures. In these experiments, no heat was added during regeneration, and we used nitrogen instead of steam to provide the CO_2 partial pressure swing. Nevertheless, the TGA tests demonstrated long sorbent life and durability over 1,644 adsoprion/desorption cycles. We maintained 10 min full cycle time (3 min adsorption, 7 min regeneration), showing that very fast rates are achievable. Figure 2 shows the comparison of adsorption uptake curves at various temperatures. At higher temperatures the rate of adsorption is higher and the sorbent reaches the equilibrium quickly.

Figure 3 shows the temperature dependence of adsorption capacity at low CO_2 partial pressures (P_{CO2} = 9.7 psia). The heat of adsorption is calculated as 5.4 kcal/mol using Henry's Law approximation for the adsorption equilibrium.

Figure 1. Long duration cycling tests at the TGA. P_{CO2} = 9.7 psia

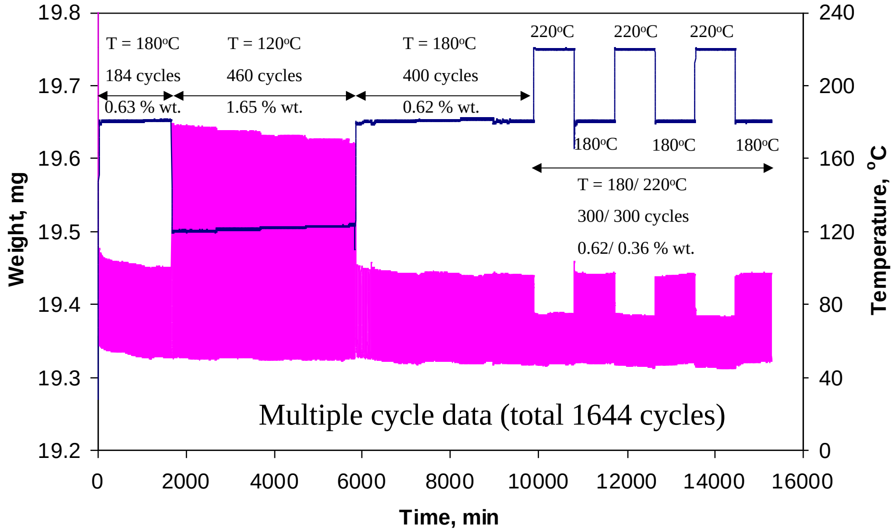


Figure 2. CO_2 adsorption uptake curves at different temperatures. P_{CO2} = 9.7 psia

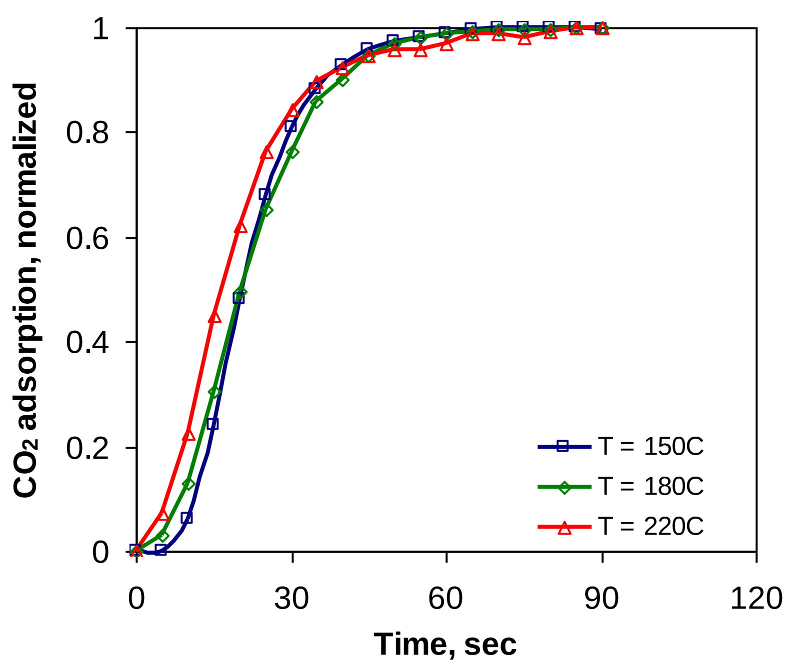
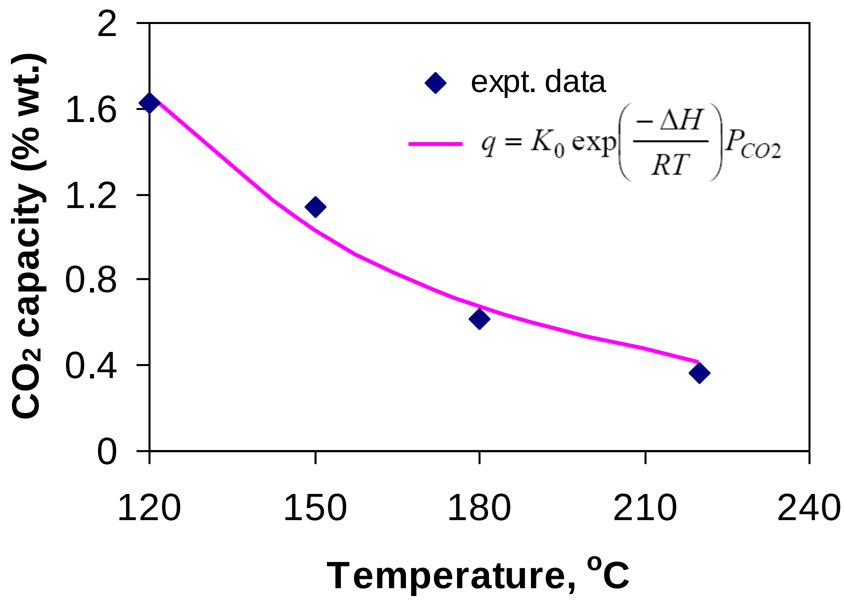


Figure 3. CO_2 adsorption capacity at different temperatures. P_{CO2} = 9.7 psia



SUMMARY

TDA has developed a novel sorbent that removes CO_2 via physical adsorption (5.4 kcal per mol of CO_2 adsorbed), which is much lower than amine-based solvents and chemical absorbents. We carried out bench-scale evaluations of TDA's CO_2 sorbent in both fixed-bed adsorber and a thermo gravimetric analyzer (TGA). The sorbent showed stable performance in both these systems under simulated synthesis gas (44 cycles in the bench-scale flow system and over 1,644 cycles in the TGA). The sorbent achieved high CO_2 capacity, high removal efficiency and excellent durability. We also studied the effect of operating parameters like temperature, pressure, and cycle time on the sorbent performance. The results from our sorbent evaluations will be presented at the meeting.

REFERENCES

1. Annual Energy Outlook 2007, Report #:DOE/EIA-0383(2007)
2. Tonks, Bill (2007). "CBM/CMM/AMM/VAM Mitigation, Past-Present-Future" presented at *1st Annual US EPA CMOP CMM Conference* St. Louis, MO 25-27 September(http://www.epa.gov/cmop/docs/cmm_conference_sep07/camco_international_2007.pdf)