A System for Capturing CO$_2$ from a Combustion Exhaust – Feasibility Research

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ABSTRACT

A Particle Gas Absorber system designed to remove CO$_2$ from a rapidly flowing exhaust gas is described. Absorbent particles are evenly dispersed at the top of a duct to fall through and absorb CO$_2$ from a gas flowing through the duct. The CO$_2$ diffuses first through the gas and then through the water on the particle to react and form a product stored on the particle. The particles are sized to absorb CO$_2$ on the appropriate time scales, and are collected and washed outside the duct for recycling. The diffusion processes are modeled and the appropriate particle chemistry is developed and measured. Static absorption experiments are performed to measure gas absorption rates. Process economics is estimated, future work is described, and overall feasibility assessed.

Keywords: CO$_2$, capture, combustion, global, warming

1 PARTICLE GAS ABSORBER SYSTEM

A dispersed-particle gas absorber (PGA) is being developed to remove CO$_2$ from industrial and power generating combustion exhausts. The basic configuration of the PGA is shown in Fig. 1. Large numbers of evenly spaced, small particles falling through an exhaust gas
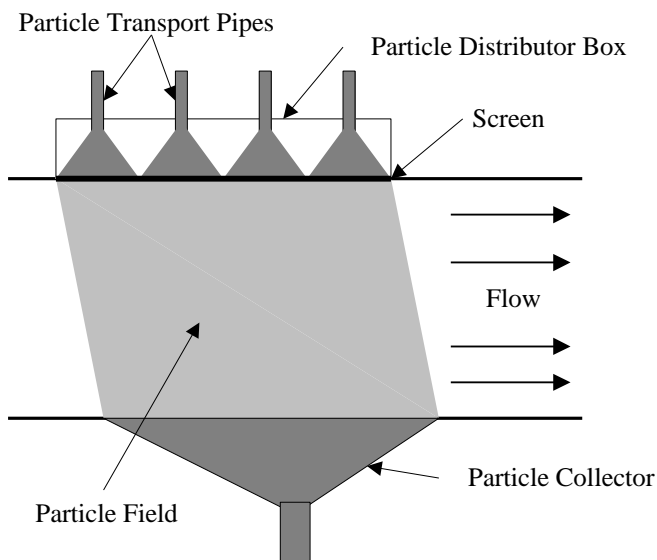

Figure 1. Schematic of Particle Gas (PGA) Absorber System

stream are used to efficiently absorb CO$_2$ from the gas.

This technique allows large gas/solid surface contact area for diffusion with only a small associated pressure drop in the flow. The particles are liquid/solid combinations that can absorb large amounts of CO$_2$ from the gas such that the CO$_2$ can be reversibly removed externally from the collected particles and reused in a continuous low-cost process. The particles and the particle field are sized such that there is an appropriate residence time to absorb most of the CO$_2$ from the warm gas. Critical issues are particle recycling and distribution in the gas flow, absorption effectiveness, process sensitivity to real conditions, installation, and overall economics.

An engineering prediction for the PGA has been derived from calculations of the performance of related droplet heat exchangers. Droplet heat exchangers (DHXs) are well developed, both analytically and experimentally, using droplets instead of particles [1]. The droplet speeds, volume and mass loadings are similar to that proposed here. Droplet heat exchangers have an effectiveness (ratio of actual heat transfer rate to thermodynamic maximum heat transfer rate) of 0.9 - 0.95 [2]. The disadvantages of droplet heat exchangers include fluid loss and the need to make droplet injectors that form droplets within a very narrow size range. Heat (scalar) transfer is very efficient because of the very large total direct contact area. DHXs are well established experimentally, but not used commercially.

The basic design parameters of the system include the particle mass loading, the particle size, residence time, and the particle CO$_2$ storage capacity. A PGA is a fundamentally different device compared with a droplet heat exchanger in that the PGA particles are a total sink for the absorbed gas, whereas the droplets in the heat exchanger can only store heat based on the steadily decreasing temperature difference between the droplets and the flow as the heat exchange proceeds. This means that the PGA can absorb 100% of the target gas from the flow, whereas a droplet heat exchanger can at best absorb only a major fraction of the heat from the flow.

The particle size in the PGA is determined both by the CO$_2$ absorption/reaction rate and the speed at which the particles fall through the duct. For instance, a 0.1 mm diameter spherical particle of specific gravity 2.0 falling through air at standard temperature and pressure, has a terminal velocity of approximately 50 cm/s and would fall across a 2 m exhaust duct in about 4 s. The particles must then absorb their capacity of CO$_2$ in this time period.

Furthermore, the length of the particle field and the number of particles in that field volume must be sufficient to absorb all of the CO_2 in a time defined by the length of the field divided by the flow velocity.

A 5% CO_2 loading in a 2m x 2m power plant exhaust duct at 100^oC implies 10^{20} CO_2 molecules in a liter of the exhaust gas. For a 1 m/s flow speed, this implies a CO_2 mass flow of 290 g/s that must be absorbed in the particles. For a particle volume loading of 1%, and a particle field length of 1 m, there will be a total mass of 20 kg of particles in the duct at any one time. At a terminal velocity 50 cm/s, there will be a 0.5 m^3/s volume flow rate of the particle field, and thus a 10 kg/s mass flow rate of particles, such that the absorbed CO_2 adds about 3% to the particle mass, which is readily accomplished.

The PGA system has many other advantages aside from minimal flow effects. It has a high flow velocity dynamic range, due to the ability for changing the mass fraction of seeded particles. Particles of uniform size are used, so no particles are lost to the air flow. In terms of chemistry it offers direct transfer of the CO_2 to a water soluble form that can readily be transformed into a commercially useful product. The particle material and chemistry have been chosen to tolerate the warm, corrosive environments typical of combustion exhausts. Water vapor and condensation are allowed and compatible with the particles, since the process needs water vapor to function properly. The particle washing process for extraction of the CO_2 product also removes other pollutants from the gas stream as well as contaminants from the particles. The process may be adaptable to simultaneous removal of pollutants other than CO_2. A major practical advantage is the ease of retrofitting the proposed absorber system, compared to competing technologies.

2   THE PARTICLES

Particle development is being performed by the Kumar group at the University of Connecticut. The target problem is to develop water-based chemistry that can be performed on small particles with the capability of absorbing large amounts of CO_2 in short (1-5 seconds) periods. The fundamental reaction for transforming CO_2 in the gas to a storable form is to create carbonic acid, which can then be reacted to form a variety of products that have commercial use. This process is normally slow but has been greatly accelerated; a variety of chemistries have been explored Another constraint is the thermal stability of the process – it must tolerate gas temperatures approaching 100^oC for short durations, the chemical environment of the exhaust stream, and retain the conversion efficiency over cycles of use.

The chemistry of the particles is being characterized by physical, chemical and biochemical approaches. The CO_2 conversion is followed either by monitoring the partial pressure of the gas with a pressure transducer, or by monitoring the pH of the medium after equilibration of samples at known partial pressures of CO_2. Calibration

graphs are being constructed and Michaelis constant (K_m), and maximum reaction velocity (V_{max}), are being estimated for specific reaction conditions (pH, temperature, and gas composition) by plotting initial reaction velocities directly. Lineweaver-Burke plots of these data give equal weight to all results (low and high substrate concentrations) and explicitly indicate diffusional limitations.

3   PROCESS MODELING

Modeling of the process is being carried out together with Prof. Yavuzkurt of the Pennsylvania State University department of mechanical and nuclear engineering. Basic analytical modeling of the CO_2 transfer process. Initial modeling efforts centered on identifying all of the diffusion processes, and then determining the time constants of each process to discover the rate-controlling process(es). The processes are illustrated in Fig. 2, and, in
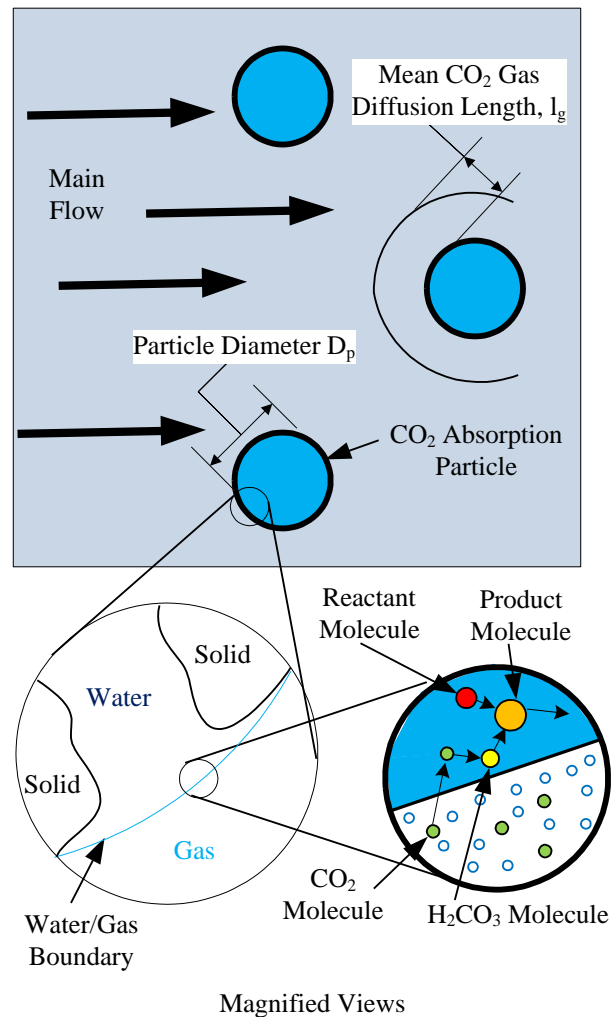

Magnified Views

Figure 2. CO_2 Absorption Modeling Physical Schematic

sequence, are 1) Diffusion of CO_2 through the bulk gas to the particle, 2) Diffusion of the CO_2 across the gas/water boundary and into solution, 3) Diffusion of CO_2 through the

water to the reaction site, 4) Reaction of the CO_2 to form carbonic acid, 5) Diffusion of the other reactant to the carbonic acid, 6) Reaction of the reactants, and 7) Diffusion of the reaction product into the bulk water.

The particles are small enough that the Reynolds Number , Re_d, is small and no boundary layers are formed. A typical particle is in the wake flow of upstream particles, but this effect is ignored to first order, and each individual particle is assumed to be exposed to steady gas cross flow. Mass transfer rate through the gas is controlled defined by the Sherwood numbers, calculated as a function of flow velocity and particle density. Experimental diffusion coefficients were determined from the literature. Based on these calculations a particle size was determined so that the CO_2 absorption could take place on the required time scale.

4 CO_2 CAPTURE EXPERIMENTS

Initial experiments were conducted in a stationary pure CO_2 gas. A quartz-tube vacuum chamber was fabricated that allows visualization of the particle injection/seeding and the transit of the particles through the volume. Total vacuum tightness allows evacuation to very low pressures allowing pure-gas testing at arbitrary subatmospheric pressure and eliminates the possibility of contamination during tests. CO_2 absorption in water was tested first, demonstrating the performance of the pressure apparatus in a horizontal configuration for maximum water surface area. Bubbling pure CO_2 through distilled water at 1 atm and room temperature gave an asymptotic pH of 4.0.

The apparatus was then reconfigured as shown in Fig. 3 to perform spray/seeding experiments. The fill pressure is monitored by a diaphragm pressure gauge that accurately measures pressure independent of gas composition. Particle/water/reactant mixtures are be fed or sprayed into the chamber, collecting the particle/water at the bottom. At the end of the experiment, the fluid can be drained to be tested for pH.

Experiments are being done using pure CO_2 at a range of pressures, followed by confirming experiments in mixtures of CO_2 in air. An optical CO_2 monitor is used to measure the CO_2 concentration in the presence of other gases.

Initially, the gas is pure CO_2 and, after the particles are injected, water vapor. The initial CO_2 concentration is set by the pressure in the chamber. The typical concentration of CO_2 in a combustion gas is 5-10%. This

5 PROCESS ECONOMICS

The particles are the most expensive components of the system; operation is inexpensive and the impact on the exhaust flow is small. If the particles and the reaction chemistry can be kept active for 10 hours of continuous use the estimated process cost will be reduced to 0.3-0.4 cents per mole of CO_2. A 1000 MW coal-fired power station typically produces about 7 million metric tonnes of CO_2 per year. This is equal to 1.6×10^{11} moles per year. The total cost per year is then 480 million dollars per year or $68 per tonne. This cost is considerably lower than the currently projected sequestration cost of $200 per tonne [3] There is potential to reduce this figure by a factor 10 by producing the particles in large quantities.
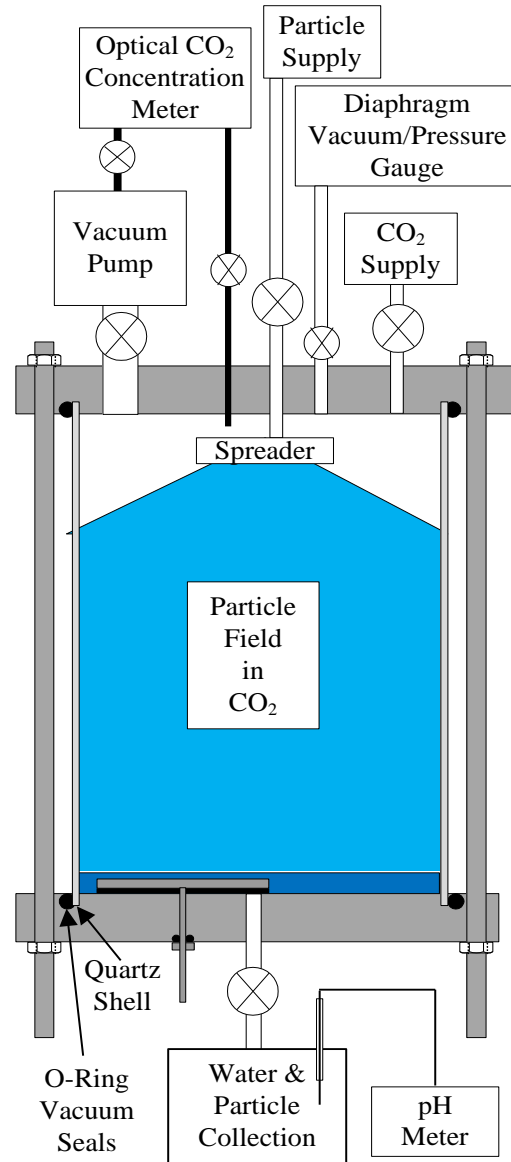

Figure 3. CO_2 absortion measurement apparatus schematic.

Capital cost of the facility is low. The most expensive part is the seeding mechanism, which may be simple. The particles are gravity drained, washed, and spun to remove the proper amount of water, then pumped to the top of the duct for reuse. The system can be installed as a retrofit, requiring no additional major facilities. Furthermore, the system probably can be installed during operation, avoiding a major and expensive shutdown.

6 FUTURE WORK

Near-term future work will complete particle development and static experiments, fabricate and perform flow experiments, and perform a final feasibility assessment.

For the flow experiments, a small burner system and exhaust cooling will be fabricated to supply a variably warm, CO_2 laden flow for absorber tests. The pre-treated, properly sized particles will then be seeded into the duct, and collected at the bottom. The overall gas flow rate will control residence time by adjusting gas feed rates. Air flow will be monitored with a flow meter, and average CO_2 transfer coefficients will be obtained. During these experiments particle field aerodynamic behavior will be observed by taking pictures of the illuminated flow field through a transparent duct side. The device operating range will be explored. Gas and particle mass flow rate must be matched so that most of the CO_2 gas contained in the flow is removed by the particles. There is a tradeoff between final CO_2 removal efficiency and overdosing of the flow with particles. Critical aspects of the experiments will be the fraction of CO_2 captured as a function of both the mass flow of gas and the mass flow of particles.

Long-term future work includes designing and building a prototype device. Detailed modeling of the mass transfer and CO_2 absorption processes will be carried out, comparing model results with experiments and using them to guide scale-up design.

7 CONCLUSIONS

Modeling, economic analysis, and preliminary experiments indicate that the Particle Gas Absorber concept will be a practical means for removing CO_2 from combustion exhausts. The particle mass loading can be adjusted to absorb all of the CO_2, and the particle size and chemistry can be adjusted to absorb the CO_2 during the transit time across the duct enclosing the flowing gas. Furthermore, the particles can be designed to tolerate the harsh conditions of a combustion exhaust. Detailed work remains to be done on thermal stability, particle recycling and overall efficiency.

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