PROJECT facts

U.S. Department of Energy
Office of Fossil Energy
National Energy Technology Laboratory

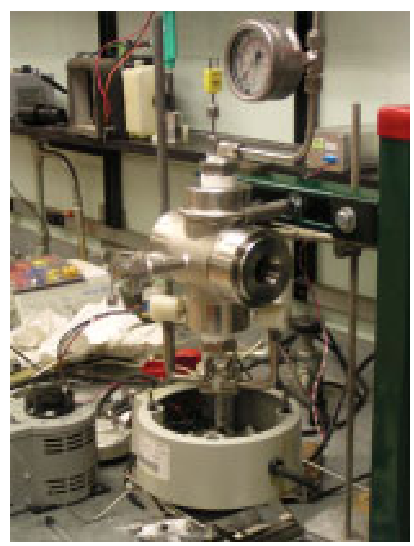
Sequestration

04/2004

Laboratory Investigations in Support of Carbon Dioxide-Limestone Sequestration in the Ocean

Background

Many approaches have been proposed for the sequestration of CO_2. One idea, which has received much consideration, is that of storing CO_2 in the ocean. However, since liquid CO_2 is less dense than water and poorly miscible with water, the CO_2 must be injected at sufficient depth, so it will not buoy upward to approximately 500 m depth, where it would flash into vapor and reemerge into the atmosphere. Furthermore, when CO_2 dissolves in water it forms carbonic acid, which lowers the pH of seawater, and may have an adverse effect on oceanic biota. To circumvent these problems, the UML researchers proposed to inject into the ocean not pure liquid CO_2, but an emulsion of CO_2 in water stabilized by limestone ($CaCO_3$) particles. The emulsion is heavier than seawater, hence it will sink deeper from the injection point rather than buoy upward. Secondly, the $CaCO_3$ coated CO_2 droplets will not acidify the seawater. In the first year of the NETL sponsored contract, the UML researchers found that, under proper conditions, liquid CO_2 will form an emulsion in water in the presence of powdered limestone in which the globules of CO_2 are denser than water. In the second year of the contractual period the UML researchers would like to optimize the conditions for globule formation, including CO_2 to $CaCO_3$ ratio, and $CaCO_3$ particle size, as well as globule stability over long periods. In the third year extension of the contract, the effect of impurities and ion strength on globule formation will be investigated, as well as the possibility of using other particles than $CaCO_3$ for globule formation, including fly ash and various minerals. The stability of globules will also be investigated in the NETL water tunnel facility at PETC. Data collected during this phase will facilitate the development of modeling for future scaleup work.



The high pressure batch reactor in which CO_2-in-water emulsions are formed stabilized by powdered limestone particles.

Primary Project Goal

The general objective of this work is to establish a database to enable the evaluation of an improved process for the deep water ocean sequestration of CO_2. The process forms globules of liquid CO_2 in water, with the globules being stabilized by particles of limestone at the CO_2/water interface.

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PARTNERS

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COST

Total Project Value
$577,518

DOE/Non-DOE Share
$481,551 / $95,967

Benefits

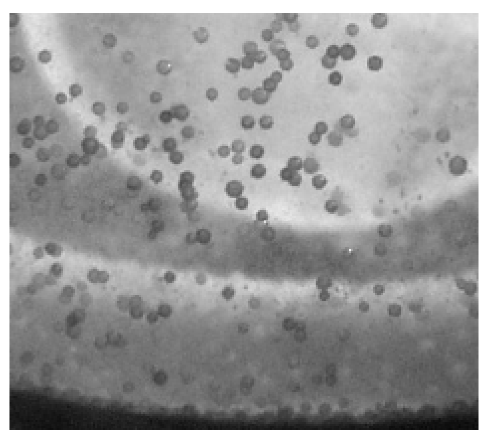
Concerns about the contribution of greenhouse gas emissions to global warming have led to the study of ways to capture and sequester CO_2 at major emitting sources (e.g. fossil fueled power plants and industrial boilers) to prevent its emission into the atmosphere. One potential sink for CO_2 are the oceans of the world, with almost unlimited capacity to sequester CO_2. However, dissolving CO_2 in seawater lowers its pH, which may have adverse effects on aquatic organisms. If this project is successful, it could provide a method for ocean sequestration of CO_2 that would avoid this problem, thus making it possible to continue the use of cheap and abundant coal and other fossil fuels until other non-CO_2 emitting energy sources become available.

Objectives

- To construct a batch high-pressure reactor in which CO_2, water, and finely ground limestone will be mixed at elevated pressure.
- To analyze emulsions in-situ using light microscopy and light scattering to determine their structural properties, the size of the droplets and $CaCO_3$ particles that stabilize the emulsions, hydrate formation, and other significant properties.
- To vary initial conditions (pressure, temperature, ingredients, water type, particle size, etc.) to determine the effects on emulsion physical and chemical characteristics.
- After successful completion of batch experiments, to convert the reactor into a flow system in which liquid CO_2 and pulverized limestone can be fed continuously and thoroughly mixed to form an emulsion.
- To use the flow system to investigate the physical and chemical characteristics of the emulsions as a function of time while varying initial conditions.
- To analyze the data to report findings on observed relationships between measured characteristics and operating conditions.
- To perform a simple economic analysis of the costs associated with the process, which will reflect the amounts and costs of raw materials (limestone or other particles) and the energy required to pulverize, mix and transport the emulsion to the deep ocean, expressed as the cost of sequestering one ton of CO_2 in the ocean.

Accomplishments

A high-pressure batch reactor with a view window has been constructed. This reactor was used to conduct a wide range of tests using various proportions of liquid CO_2, water, and pulverized limestone to form emulsions of CO_2 droplets in water stabilized by $CaCO_3$ particles. After thorough mixing of the ingredients, a stable emulsion forms with globules consisting of an inner core of liquid CO_2 coated with a sheath of $CaCO_3$ particles dispersed in water. Using limestone particles with a size range of 6-13 ìm and the proper stirring conditions, globules with diameters of 100-200 ìm were formed which were denser than water and sank to the bottom of the high pressure reactor. The globules were observed for many hours and appear to be stable. Furthermore, the water in the reactor had a pH in the range of 7-10 compared to a pH of 3-4 for carbonic acid. It was also demonstrated that artificial seawater (3.5% NaCl solution) can be used instead of deionized water to form a stable emulsion. It has been estimated that about 0.5 to 0.75 tons of pulverized limestone is required per ton of CO_2 for stable emulsion formation. The construction of the flow reactor has been commenced in which the conditions for stable emulsion formation can be further studied, and the long time stability of the formed globules can be investigated.



A close-up view of the CO_2 globules coated with a sheath of limestone particles. Globules are settling out of suspension.