PROJECT facts

Carbon Sequestration

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

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

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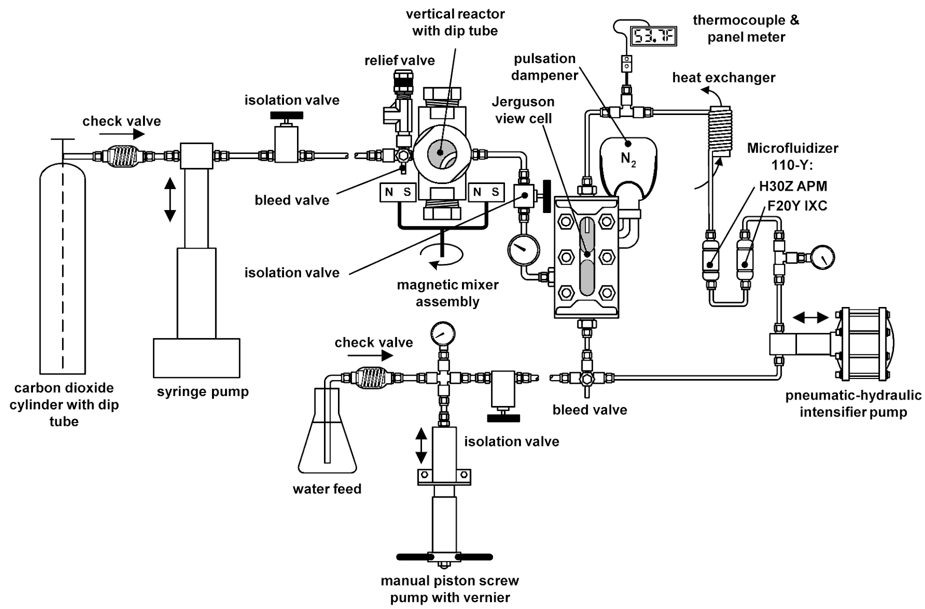
Background

Many approaches have been proposed for the sequestration of carbon dioxide (CO_2); one being considered is storing CO_2 in the ocean. However, because liquid CO_2 is less dense than water and poorly miscible with water, it must be injected at sufficient depth so it will not buoy upward. At approximately 500 meters depth, 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 adverse effects on marine biota. To circumvent these problems, researchers proposed to inject into the ocean an emulsion of CO_2 in water stabilized by limestone (calcium carbonate, $CaCO_3$) particles. The emulsion is heavier than seawater, and sinks deeper from the injection point rather than buoying upward. The $CaCO_3$-coated CO_2 droplets will not acidify the seawater.

Description

Under specific conditions, liquid CO_2 will form an emulsion in water in the presence of powdered limestone, and the globules of CO_2 are denser than water. Researchers plan to optimize the conditions for globule formation, including the CO_2 to $CaCO_3$ ratio and $CaCO_3$ particle size, as well as globule stability over long periods. They also plan to study the effect of impurities and ion strength on globule formation, as well as the possibility of using particles other than $CaCO_3$ for globule formation, including fly ash and various minerals. The stability of globules will also be investigated in the National Energy Technology Laboratory's High Pressure Water Tunnel Facility. Data collected will facilitate model development for future scale-up work.



Schematic of the microfluidizer with auxiliary feed mechanism and Jerguson view cell.

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 limestone particles at the CO_2/water interface.

Objectives

The research objectives for this project are to:

- Construct a batch high-pressure reactor to mix CO_2, water, and fine limestone mixed at elevated pressure.
- Analyze emulsions in-situ using light microscopy and light scattering to determine their structural properties.

- Determine the effects of varied initial conditions (pressure, temperature, ingredients, water type, particle size, etc.) on an emulsion's physical and chemical characteristics.

- Convert reactor into a flow system in which liquid CO_2 and pulverized limestone can be fed continuously and thoroughly mixed to form a stable emulsion.

- Use the flow system to investigate the physical and chemical characteristics of the emulsions as a function of time while varying initial conditions.

- Analyze the data to report findings on observed relationships between measured characteristics and operating conditions.

- Perform a simple economic analysis of the costs associated with the process, expressed as the cost of sequestering one ton of CO_2 in the ocean. Factors will include: amounts and costs of raw materials and the energy required to pulverize, mix and transport the emulsion to the deep ocean.

PERFORMANCE PERIOD

07/09/2002 to 03/31/2008

COST

Total Project Value
$853,030

DOE/Non-DOE Share
$668,208 / $184,822

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 powerplants and industrial boilers) to prevent release to the atmosphere. One potential sink for CO_2 is the oceans of the world, with almost unlimited capacity to sequester CO_2. If this project is successful, it could provide a method for ocean sequestration of CO_2, 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.

Accomplishments

A high-pressure batch reactor with view window was constructed and 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, a stable emulsion was formed, with globules consisting of an inner core of liquid CO_2 coated with a sheath of $CaCO_3$

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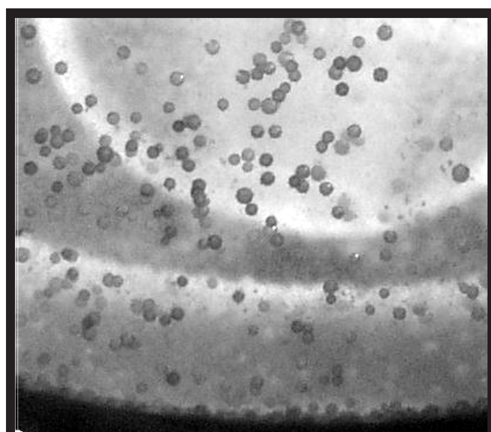
particles dispersed in water. Using limestone particles with a size range of 6-13 microns and the proper stirring conditions, globules of 100-200 microns diameter were formed; denser than water, they sank to the bottom of the reactor. The globules were observed for more than 24 hours and appeared to be stable. Water in the reactor had a pH of 7-10, compared to a pH of 3-4 for carbonic acid. Research also demonstrated that artificial seawater (3.5 percent sodium chloride solution) can be used instead of de-ionized water to form a stable emulsion. Researchers estimated that about 0.5 to 0.75 tons of pulverized limestone would be required per ton of CO_2 to form stable emulsions.

The microfluidizer was successfully used to create liquid CO_2-in-water and water-in-liquid carbon dioxide emulsions using fine particles as stabilizers. For CO_2-in-water emulsions, hydrophilic particles of finely milled $CaCO_3$ were used as stabilizers; for water-in-liquid carbon dioxide emulsions, hydrophobic pulverized Teflon particles were used as stabilizers. The $CaCO_3$-sheathed CO_2 globules were lighter than water, and floated on top of the water column – a process called creaming. The Teflon-sheathed water globules were heavier than liquid carbon dioxide, and sank to the bottom of the liquid CO_2 column – a process called sedimentation.



Close-up view of CO_2 globules coated with a sheath of limestone particles and settling out of suspension.

Planned Activities

Remaining experimentation will focus on operation of the microfluidizer to investigate factors limiting production of stable micro-emulsions. It may be possible to create a homogeneous micro-emulsion without creaming and sedimentation by increasing the shear force imparted by the microfluidizer on the interacting liquids and decreasing the stabilizing particle size. This may be necessary to ensure that the micro-emulsions penetrate the pore spaces of geologic formations for geologic sequestration applications. A final report will be submitted after completion of this research in March 2008.