

# PROJECT facts

U.S. DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
NATIONAL ENERGY TECHNOLOGY LABORATORY

Carbon Sequestration

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## A NOVEL APPROACH TO MINERAL CARBONATION: ENHANCING CARBONATION WHILE AVOIDING MINERAL PRETREATMENT PROCESS COST

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### Background

Carbonation of the widely occurring minerals of the olivine group, such as forsterite ( $\text{Mg}_2\text{SiO}_4$ ), is a potential large-scale sequestration process that converts  $\text{CO}_2$  into the environmentally benign mineral magnesite ( $\text{MgCO}_3$ ). Because the process is exothermic, it inherently offers low-cost potential. Enhancing carbonation reactivity is the key to economic viability. Previous studies at the U.S. DOE Albany Research Center (ARC) established that aqueous-solution carbonation using supercritical  $\text{CO}_2$  is a promising process; even without olivine activation, 30–50 percent carbonation has been achieved in an hour. Mechanical activation by attrition accelerated the carbonation process to an industrial timescale (near completion in less than an hour) at reduced pressure and temperature. However, the activation cost is too high to be economical and lower-cost pretreatment options are needed. The Arizona State University Center for Solid State Science proposes a novel approach that offers the potential to dramatically enhance carbonation reactivity while bypassing any pretreatment/activation.

Mechanistic investigations have shown that robust silica-rich layers form on an olivine surface during carbonation. As carbonation proceeds, these passivating layers thicken, fracture, and eventually exfoliate, exposing fresh olivine surfaces. Particle-particle and particle-wall collisions within the slurry stream can dramatically impact both the exfoliation rate and the extent of carbonation. Order of magnitude increases in the extent of carbonation have been observed for different flow systems. In order to identify key parameters that can enhance carbonation, it is proposed to explore exfoliation mechanisms and their relationship to enhanced carbonation using three innovative approaches:



## **PARTNERS**

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Project Team

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## **PERFORMANCE PERIOD**

06/22/2004 to 06/21/2007

- Carry out multiphase hydrodynamics fluid simulations and experimental investigations to elucidate key fluid-flow parameters that facilitate the slurry interactions that enhance exfoliation.
- Chemical studies to establish the potential for controlling passivating layer effectiveness and exfoliation rate by adjusting aqueous cation size (e.g., Li<sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>).
- Sonic investigations to elucidate the potential that controlled sonication offers to enhance exfoliation and particle cracking.

Once the key parameters for each approach are identified, they will be integrated to evaluate their combined potential to synergistically enhance exfoliation and carbonation. The above studies will be complemented by detailed morphological, structural, and compositional investigations of their intermediate and final reaction products down to the nanoscale. These studies will be further integrated with advanced computational modeling of key phenomena to develop an atomic-level understanding of the mechanisms that govern carbonation reactivity and exfoliation.

## **Primary Project Goal**

The primary goal is to develop the understanding needed to engineer a new low-cost mineral carbonation process that avoids the cost of pretreatment and/or activation.

## **Objectives**

Explore and assess the effectiveness of novel, low-cost pathways to enhance olivine passivating layer exfoliation and carbonation.

## **Benefits**

Mineral sequestration processes have the potential to permanently dispose of CO<sub>2</sub> in geologically stable mineral carbonate rocks that will not require continuous monitoring, which is required with many other CO<sub>2</sub> sequestration technologies. The technology to be developed under this proposal will hasten the natural mechanism of turning CO<sub>2</sub> into a solid. The solidification process could be accomplished in less

than an hour rather than in hundreds of thousands of years via natural mineral weathering. Methods for economically sequestering CO<sub>2</sub> will allow for continued use of our abundant coal reserves.

## Accomplishments

- Multi-phase slurry-flow studies have been integrated with multiphase hydrodynamic simulations to identify key parameters that control exfoliation and carbonation of feedstock particles. Simulations indicate that wall roughness and particle size distribution can significantly affect cross-stream transport, particle-particle collisions, and carbonation.
- A detailed systematic search for alternative aqueous solution buffers has led to the discovery that 5.5M KHCO<sub>3</sub> produces nearly twice the extent of carbonation as does the standard 0.64M NaHCO<sub>3</sub> + 1M NaCl aqueous solution, significantly enhancing the efficiency of the one-step “feedstock dissolution + aqueous carbonation” process.
- Mechanistic investigations suggest that silica-rich passivating layers (PL) formed via incongruent dissolution possess significant permeability to key reaction species during carbonation. Exfoliation behavior and structural morphology of the PL vary greatly, depending on the specific choice of halide salt and alkaline bicarbonate buffer species.
- Large-scale molecular dynamics simulations of the olivine PL predict a continuous transition zone that extends from the bulk olivine solid through the amorphous PL-solution interface and a “glassy” metastable PL nanostructure that possesses significant permeability for reactant species.
- A sonication system was developed and used to explore the effects of sonication on the extent of carbonation as a function of the weight percentage olivine present, particle size distribution, volume fraction of aqueous solution present, and sonication power, time, temperature, and CO<sub>2</sub> pressure. None of the conditions investigated to date significantly enhanced carbonation.

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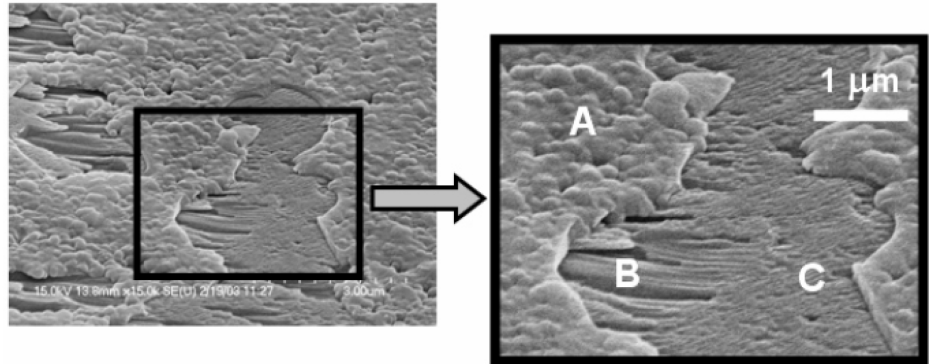
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**FIGURE 1**

*Scanning electron micrograph showing silica-rich passivating layer exfoliation. A) the passivating layer; B) a recently fractured and exfoliated region exposing part of the olivine particle core; C) a new passivating layer beginning to grow in the exfoliated region.*