

## Biomineralization Processes Using Fly Ash for Carbon Sequestration

석탄회를 이용하는 탄소 격리용 생광물화 작용

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**ABSTRACT:** The objective of this study is to investigate biogeochemical processes to sequester CO<sub>2</sub> and metals utilizing metal-rich fly ash (MRFA). Microbial conversion of CO<sub>2</sub> into sparingly soluble carbonate minerals has been studied using MRFA under different pCO<sub>2</sub> and different bicarbonate concentrations. Scaling from test tube to fermentation vessels (up to 4-L) using metal-reducing bacteria and MRFA has proved successful at sequestering carbon dioxide and metals. CO<sub>2</sub> sequestration via precipitation processes using MRFA may complement the process of CO<sub>2</sub> capture from fossil fuel plants while potentially stabilizing fly ash wastes.

**Key words:** carbon sequestration, biomineralization, fly ash, metal-reducing bacteria

**요약:** 본 연구는 금속이 다량 함유된 석탄회(metal-rich fly ash, MRFA)를 이용, 이산화탄소 및 금속의 격리를 위한 생지화학적 과정을 연구하고자 한다. 이를 위해 다양한 이산화탄소 분압과 중탄산염 이온 농도의 조건 하에서 MRFA를 이용, 미생물에 의한 이산화탄소의 난용성 탄산염 광물로 전환 실험을 실시하였다. 시험관부터 4-L의 배양용기까지의 다양한 규모의 실험을 통해, 금속환원 박테리아와 MRFA를 이용하는 경우, 효과적인 이산화탄소 및 금속의 격리가 이루어지는 것으로 나타났다. MRFA를 이용하는 침전 기작을 통한 이산화탄소 격리는 화석연료를 이용하는 발전소에서 방출되는 이산화탄소의 제거 및 석탄회 폐기물의 안정화의 보완적 방법으로 유용하다.

**주요어:** 탄소격리, 생광물화 작용, 석탄회, 금속환원 박테리아

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## Introduction

It was perhaps one of the most stimulating scientific discoveries of the 20th century that the concentration of atmospheric CO<sub>2</sub> had been continuously increasing since the first observation was made in late 1950s at Mauna Loa, Hawaii. In addition to these several decade data, long-term records of atmospheric CO<sub>2</sub> through glacial-interglacial cycles were recovered from the air trapped in vertical sequence of glacial ice cores from the Antarctica. These data have led climate scientists, atmospheric chemists, oceanographers, geochemists, and a host of other diverse specialists to focus on efforts towards understanding how carbon is cycled among four large reservoirs of the continents, seawater, sediments, and the atmosphere. While understanding this carbon cycle remains the main topic in many studies, the consensus is that increases in atmospheric CO<sub>2</sub> by the human-related activities will play a significant role in global climate change in the near future. Energy Information Administration of the Department of Energy (DOE) estimates the atmospheric greenhouse gas releases may exceed 8 billion metric tons by the year 2010 heightening its international environmental concerns. Carbon dioxide is known to dominate the greenhouse gases, accounting for about 85% of the emissions (Kane and Klein, 1997), the majority of which results from the use of fossil fuels. With viable replacement of fossil fuels remaining decades away, alternatives to reducing the impacts of atmospheric CO<sub>2</sub> accumulations will surely include carbon sequestration and carbon management. Current costs for carbon fixation scenarios range from approximately \$60-500 per ton of CO<sub>2</sub> captured plus additional costs for transport and disposal (\$4-600/t C) (Riemer and Ormerod, 1995). We need cost effective carbon sequestration technologies coupled with very low transport and disposal costs or more preferably, the derivation of useable products.

Biological uptake via reforestation and soil formation certainly represents low costs and known technologies. Unfortunately, complete reforesta-

tion of available areas may sequester CO<sub>2</sub> less than 20% of the 1990 emissions (Riemer and Ormerod, 1995). Therefore, terrestrial primary production may be insufficient to resolve the problem. Though bio-fuels represent a sustainable option, there are cost penalties plus emissions of greenhouse gases, such as CH<sub>4</sub> and N<sub>2</sub>O (Bachu *et al.*, 1994). While injection of CO<sub>2</sub> into local geologic formations or sea floors may be a reasonable component for our carbon management strategy (Herzog *et al.*, 1991; Bachu *et al.*, 1994), Riemer and Ormerod (1995) suggested that deep ocean injection may not be immediately applicable.

Many species of microorganisms, mainly anaerobic metal-reducing bacteria (MRB), are capable of reducing amorphous and crystalline Fe oxides (Zhang *et al.*, 1997; Liu *et al.*, 1997; Roh and Moon, 2000, 2001a, and 2001b; Roh *et al.*, 2001a, 2002, and 2003). Anaerobic MRB precipitated magnetite (Fe<sub>3</sub>O<sub>4</sub>), siderite (FeCO<sub>3</sub>), vivianite [Fe<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub> · 2H<sub>2</sub>O], or sulfide (FeS) (Roh *et al.*, 2002, 2003). Recent studies demonstrated that partial pressures of CO<sub>2</sub> and ionic species composition in aqueous media exhibited profound influences on the type of minerals precipitated in anaerobic microbial cultures (Roh and Moon, 2001a; Roh *et al.*, 2003). The formation of siderite was favored in reducing environments and high CO<sub>2</sub> partial pressure (Zhang *et al.*, 1997; Roh and Moon, 2001; Roh *et al.*, 2003). The predominant minerals precipitated under a nitrogen atmosphere was magnetite. In the presence of 20% headspace CO<sub>2</sub>, a mixture of magnetite and iron rich carbonates such as siderite was formed. It has been reported that calcite (CaCO<sub>3</sub>) and magnesite (MgCO<sub>3</sub>) may be precipitated by bacteria, algae and yeast (Thompson and Ferris, 1990; Cicerone *et al.*, 1999). Determining the potential importance of the biogeochemical processes on carbonate mineral precipitation and gaining a fundamental understanding of the controlling factors, rate and extent of carbonate mineral precipitation will significantly advance our understanding of carbon management and the science of coal

utilization.

The objective of this study is to investigate biogeochemical processes to sequester CO<sub>2</sub> and metals utilizing metal-rich fly ash (MRFA). Biogeochemical conversion of CO<sub>2</sub> into sparingly soluble carbonate minerals, such as calcite and siderite, has been studied using Fe(III)-reducing bacteria in conjunction with MRFA. This research will develop a scenario by which fly ash is stabilized into carbonate solid conglomerates that could potentially be useful as fill materials or road construction aggregates.

## Materials and Methods

### Microorganisms used for Carbon Sequestration

Biogeochemical fixation of CO<sub>2</sub> into carbonate minerals having relatively low solubilities (e.g., iron carbonate and calcium carbonate) has been studied using MRB isolated from diverse environments in conjunction with materials consisting of metal-containing fly ash. Table 1 shows thermophilic and psychrotolerant Fe(III)-reducing bac-

teria isolated from a variety of cold and hot environments such as deep marine sediments, sea water near a hydrothermal vent, and deep subsurface environments. For these experiments, thermophilic (*Thermoanaerobacter ethanolicus*, TOR-39 and C1) and psychrotolerant (*Shewanella alga*, PV-4; *Shewanella pealeana*, W3-6-1 and W3-7-1) MRB (Table 1) were used to examine biogeochemical processes for CO<sub>2</sub> sequestration using MRFA.

### Biogeochemical Conditions and Fly Ash Chemistry

Culture medium contained the following ingredients (g/L): 2.5 NaHCO<sub>3</sub>, 0.08 CaCl<sub>2</sub> · 2H<sub>2</sub>O, 1.0 NH<sub>4</sub>Cl, 0.2 MgCl<sub>2</sub> · 6H<sub>2</sub>O, 10 NaCl, 0.4 K<sub>2</sub>HPO<sub>4</sub> · 3H<sub>2</sub>O, 7.2 HEPES (hydroxyethylpiperazine-N''-2-ethanesulfonic acid), 1.0 rasazurin (0.01%), 0.5 yeast extract, and 10 trace minerals and 1 vitamin solutions (Phelps et al., 1989). No exogenous electron carrier substance (i.e., anthraquinone disulfonate) and reducing agent (i.e., cysteine) were added to the medium. Biogeo-

**Table 1.** Microbial isolates chosen for investigation for carbon sequestration

Isolates	Growth condition	Site description	Geology/sample type	Genus & species	References
TOR-39	thermophilic (40~75 °C)	Taylorville Triassic Basin, North. Virginia	shale, siltstone, & sandstone	<i>Thermo- anerobacter ethanolicus</i>	Liu <i>et al.</i> , 1997
C1	thermophilic (40~75 °C)	Piceance Basin, Wasatch Fm. West. Colorado	cemented sandstone, cross-bedded siltstone & shale	<i>Thermo- anerobacter ethanolicus</i>	Liu <i>et al.</i> , 1997
PV-4	psychrotolerant (0~37 °C)	Naha vents, Coast of Hawaii	iron-rich microbial mat assoc. with a hydrothermal vent	<i>Shewanella alga</i>	Stapleton <i>et al.</i> , 2003
W3-7-1	psychrotolerant (0~37 °C)	Deep Pacific Ocean Marine Sediments	marine sediment	<i>Shewanella pealeana</i>	Stapleton <i>et al.</i> , 2003
QYMF	alkaliphilic pH=8.0~11	Boron-rich sites at U.S. Borax mine, CA	Leachate-pond contain. high level of salt (~12%NaCl) and boric acid (2~8 g/LB) at pH 9~10	<i>Alkaliphilus transvaalensis</i>	Ye <i>et al.</i> , 2003

**Table 2.** Metal-rich fly ash investigated for carbon sequestration

Material	pH	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Mineralogy
		(%)					
TVA Johnsonville ash Chattanooga, TN	8.4	44.9	20.9	24.7	2.5	1.1	mullite, Al <sub>6</sub> Si <sub>3</sub> O <sub>15</sub> maghemite, Fe <sub>2</sub> O <sub>3</sub> Quartz, SiO <sub>2</sub>
Springeville ash Joseph city, AZ	11.4	45.9	19.1	2.9	15.0	0.9	mullite, Al <sub>6</sub> Si <sub>3</sub> O <sub>15</sub> portlandite, Ca(OH) <sub>2</sub> Quartz, SiO <sub>2</sub>

chemical precipitation of carbonate minerals was performed using MRFA (Table 2) plus MRB (Table 1) isolated from diverse environments. The MRFA was obtained from several sources selected based on mineralogical and chemical characterization (Roh *et al.*, 2001a).

Experiments were performed at 25 °C for psychrotolerant and alkaliphilic cultures (*Shewanella pealeana*, W3-7-1; *Shewanella alga*, PV-4; *Alkaliphilus transvaalensis*, QYMF), and at 65 °C for the thermophilic cultures (*Thermoanaerobacter ethanolicus*, TOR-39) (Table 1). Experiments were terminated after 30 days of incubation for psychrotolerant and alkaliphilic bacteria and after 22 days for thermophilic bacteria.

Biogeochemical processes for carbon sequestration using MRB isolated from diverse environments (Table 1) and MRFA (Table 2) were examined in the presence of different pCO<sub>2</sub> including N<sub>2</sub>, N<sub>2</sub>-CO<sub>2</sub> (99.95% N<sub>2</sub>-0.05% CO<sub>2</sub>; 95% N<sub>2</sub>-5% CO<sub>2</sub>; 80% N<sub>2</sub>-20% CO<sub>2</sub>), H<sub>2</sub>-CO<sub>2</sub> (80% H<sub>2</sub>-20% CO<sub>2</sub>), CO<sub>2</sub> (100% CO<sub>2</sub>). Experiments with pCO<sub>2</sub> (0.05%CO<sub>2</sub>) close to the atmospheric CO<sub>2</sub> content were also examined to see the influence of low CO<sub>2</sub> on carbon sequestration. In addition, the effects of bicarbonate buffer concentration on biogeochemical processes for carbon sequestration were also examined using different concentrations of HCO<sub>3</sub><sup>-</sup> buffered media (30~210 mM). The pH of the medium with MRFA was varied from 6.5 to 9.5. Hydrogen (80% H<sub>2</sub>-20% CO<sub>2</sub>), glucose (10 mM), acetate (10 mM), or lactate (10 mM) served as electron donors to examine biogeochemical sequestration of CO<sub>2</sub>.

### Geochemical and Mineralogical Characterization

To examine biogeochemical processes such as dissolution and carbonation using MRFA in the presence of different pCO<sub>2</sub> and bicarbonate buffer concentrations, supernatants and fly ash slurry were geochemically and mineralogically characterized. The redox potential (Eh) and pH values in bacterial cultures at the beginning and end of the experiments were measured at room temperature in an anaerobic chamber. The pH measurements used a combination of pH electrode and an ORION EA 920 expandable ion analyzer (Orion Research, Beverly, MA), standardized with pH buffer 7 and the appropriate buffer of either pH 4 or 10 (Roh *et al.*, 2001). Eh values were measured using platinum micro-electrodes (Microelectrodes, Inc., Londonerry, NH) (Roh *et al.*, 2001). The probe was placed directly into the sample tube and equilibrated for at least 5 minutes before recording the value. Dissolved metals including Ca, Na, Fe, and other dissolved elements in the solution with fly ash were determined by inductively coupled plasma-mass spectroscopy (ICP-MS) (Model: SCIEX, Perkin Elmer). Total carbon contents of the MRFA were determined using a carbon determinator (CR-12 dry combustion furnace, Leco, St. Joseph, MI). The mineralogy of fly ash was determined using X-ray diffractometer (Model: XDS 2000, Scintag, Inc, Sunnyvale, CA) with Co-K $\alpha$  radiation (40 kV/35 mV) and a scan rate of 2°2 $\theta$ /min. Chemical compositions of the fly ash were determined using inductively coupled plasma-optical emission spectroscopy (ICP-OES) (Model:

Optima 4000 DV, Perkin Elmer). A scanning electron microscope (SEM) (Model: JSM-35CF, JEOL Ltd., Tokyo, Japan) with energy dispersive spectrometer (EDS) was used for the analysis of fly ash particle morphology and elemental compositions.

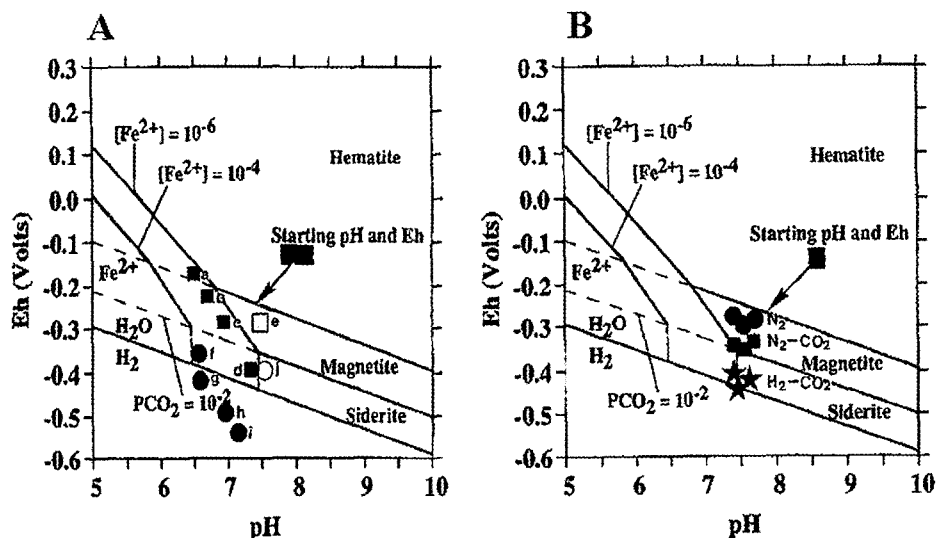
## Results and Discussion

### Solution Chemistry

Measurements of Eh and pH values were plotted on Eh-pH stability fields for hematite, magnetite, and siderite in the Fe-H<sub>2</sub>O-CO<sub>2</sub> system at 25 °C and 1 atm total pressure (Zhang *et al.*, 1997) (Fig. 1). During the growth of the Fe(III)-reducing bacteria, pH decreased from 8.0 to 6.5 and Eh decreased from ~40 mV to -550 mV (Fig. 1). Microbial processes with lactate and fly ash under a higher bicarbonate buffer (140 ~210 mM) resulted in lower Eh values than

microbial processes with a lower bicarbonate buffer (30~70 mM) (Fig. 1A), suggesting greater microbial reduction of Fe(III) in association with the increased bicarbonate buffering capacity.

Similarly, the microbial utilization of hydrogen under a H<sub>2</sub>-CO<sub>2</sub> atmosphere resulted in significantly lower Eh values (<-450 mV) than lactate utilization under a N<sub>2</sub> (~200 mV) and a N<sub>2</sub>-CO<sub>2</sub> (~300 mV) atmosphere (Fig. 1B), suggesting greater microbial reduction of Fe(III) in association with H<sub>2</sub> oxidation. The observation of microbial siderite and calcite formation using MRFA in a higher bicarbonate buffer (210 mM) and under a H<sub>2</sub>-CO<sub>2</sub> atmosphere was consistent with the Eh measurement. The presence of a H<sub>2</sub>-CO<sub>2</sub> atmosphere and the high bicarbonate buffer (210 mM) provided more reducing conditions and significant buffering capacity allowing the complete reduction of Fe(III) in MRFA in contrast to the N<sub>2</sub>/N<sub>2</sub>-CO<sub>2</sub> atmosphere and low



**Fig. 1.** Eh-pH stability fields for hematite, magnetite, and siderite in the Fe-H<sub>2</sub>O-CO<sub>2</sub> system at 25 °C and 1 atm total pressure (modified from Zhang *et al.*, 1997): (A) measured Eh and pH under different bicarbonate concentration (a: 30 mM HCO<sub>3</sub><sup>-</sup>, TOR-39; b: 70 mM HCO<sub>3</sub><sup>-</sup>, TOR-39; c: 140 mM HCO<sub>3</sub><sup>-</sup>, TOR-39; d: 210 mM HCO<sub>3</sub><sup>-</sup>, TOR-39; e: control; f: 30 mM HCO<sub>3</sub><sup>-</sup>, C1; g: 70 mM HCO<sub>3</sub><sup>-</sup>, C1; h: 140 mM HCO<sub>3</sub><sup>-</sup>, C1; i: 210 mM HCO<sub>3</sub><sup>-</sup>, C1; j: control) and (B) measured Eh and pH under different atmospheric composition including N<sub>2</sub>, N<sub>2</sub>-CO<sub>2</sub> (80% N<sub>2</sub>-20% CO<sub>2</sub>), and H<sub>2</sub>-CO<sub>2</sub> (80% H<sub>2</sub>-20% CO<sub>2</sub>).

bicarbonate buffer (30~140 mM). Thus, the Eh-pH diagram shows that carbonate minerals including calcite and siderite precipitation are likely to be facilitated by the microbial alteration of Eh and pH conditions, or both, and creating conditions of potentially localized supersaturation with respect to a mineral phase (Zhang *et al.*, 1997).

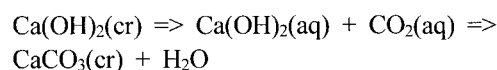
Chemical analysis of water-soluble metals in the culture medium after incubation revealed that the leaching of Ca and Fe from fly ash was significantly reduced in the presence of a H<sub>2</sub>-CO<sub>2</sub> atmosphere (Fig. 2b) and in HCO<sub>3</sub><sup>-</sup> buffered media (>140 mM) (data not shown). However, considering greater microbial reduction of Fe in association with H<sub>2</sub> and higher bicarbonate concentration, in this condition, leaching of major cations was not reduced, but leaching and active precipitation increased, therefore, we could conclusively detect reducing trend. In addition to microbially facilitated precipitation of carbonate minerals using fly ash, biogeochemical processes produced sparingly soluble carbonate minerals contributing to direct or indirect precipitation and sequestration of redox sensitive metals. This effect was likely a consequence of microbial metal reduction and the (co)precipitation of carbonate minerals in the presence of appropriate electron donors such as hydrogen, lactate, and glucose.

#### *Biogeochemical Carbon Sequestration under Different pCO<sub>2</sub> and Bicarbonate Concentrations*

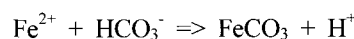
The atmospheric composition and bicarbonate buffer concentration in conjunction with biogeochemical processes exhibited profound influences on the types of minerals and the rate of carbonate mineral precipitation. Bottles containing MRB and an energy sources led to more biogeochemically facilitated precipitation of carbonate minerals with redox-labile metals.

XRD analysis (Fig. 3) showed that the biogeochemical processes induced precipitation of calcium carbonate, calcite (CaCO<sub>3</sub>), using Ca-rich Springerville fly ash under the CO<sub>2</sub> atmospheres

and with bicarbonate buffer at 60°C incubation temperature. SEM with EDX spectra also showed that biogeochemical processes caused the precipitation of calcium carbonate using Ca-rich fly ash under a H<sub>2</sub>/CO<sub>2</sub> atmosphere and a high bicarbonate buffer (210 mM) (Roh *et al.*, 2001a). No carbonate minerals formed using MRFA without bacteria. The biogeochemical processes facilitated calcite precipitation using Ca-rich fly ash or Ca-poor fly ash under a H<sub>2</sub>-CO<sub>2</sub> atmosphere and a high bicarbonate buffer (210 mM):



XRD analysis showed that increased bicarbonate buffer (210 mM HCO<sub>3</sub><sup>-</sup>) also facilitated biomineralization of siderite using Fe-rich Johnsonville fly ash (25% Fe<sub>2</sub>O<sub>3</sub>) under a N<sub>2</sub> atmosphere at 65 °C (Roh *et al.*, 2001a). SEM with EDX spectra showed microbially-facilitated precipitation of iron carbonate with the Fe-rich fly ash under a H<sub>2</sub>-CO<sub>2</sub> atmosphere (Roh *et al.*, 2001a). In environments with high bicarbonate concentrations, the microbial production of Fe(II) from Fe-rich fly ash may stimulate siderite formation:



The capacity of biogeochemical processes using Fe(III)-reducing bacteria to precipitate carbonate minerals such as calcite and siderite using MRFA creates the possibility of more effective CO<sub>2</sub> sequestration than would be possible with photosynthetic systems in alkaline ponds. In addition to microbially facilitated precipitation of carbonate minerals using fly ash, the microbial utilization of organic matter and hydrogen to produce sparingly soluble carbonate minerals may also contribute to (in)direct precipitation of redox sensitive metals in fly ash ponds.

Scale-up experiments (up to 4-L) using thermophilic and psychrophilic MRB have proved successful at sequestering carbon while using Ca

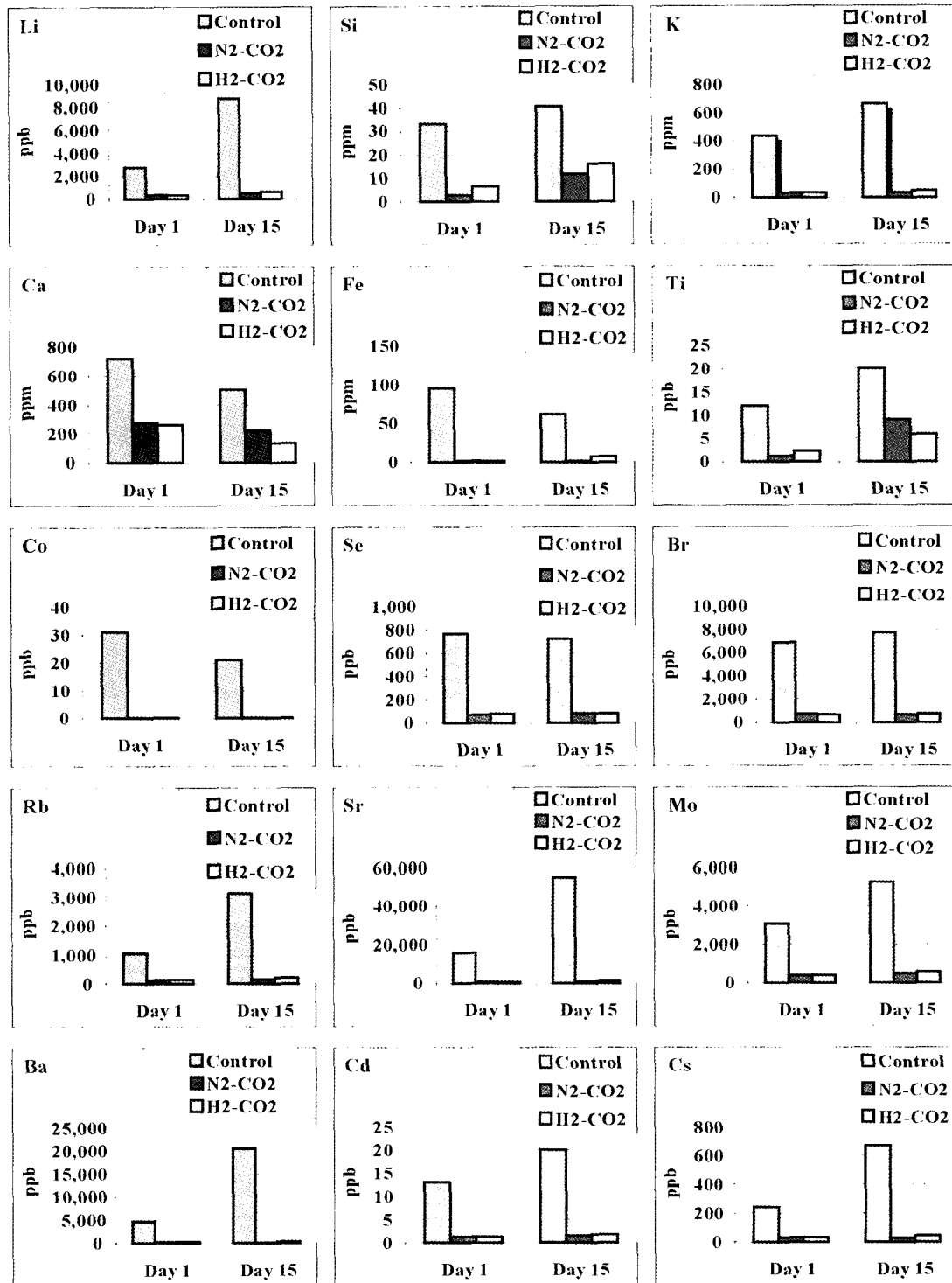
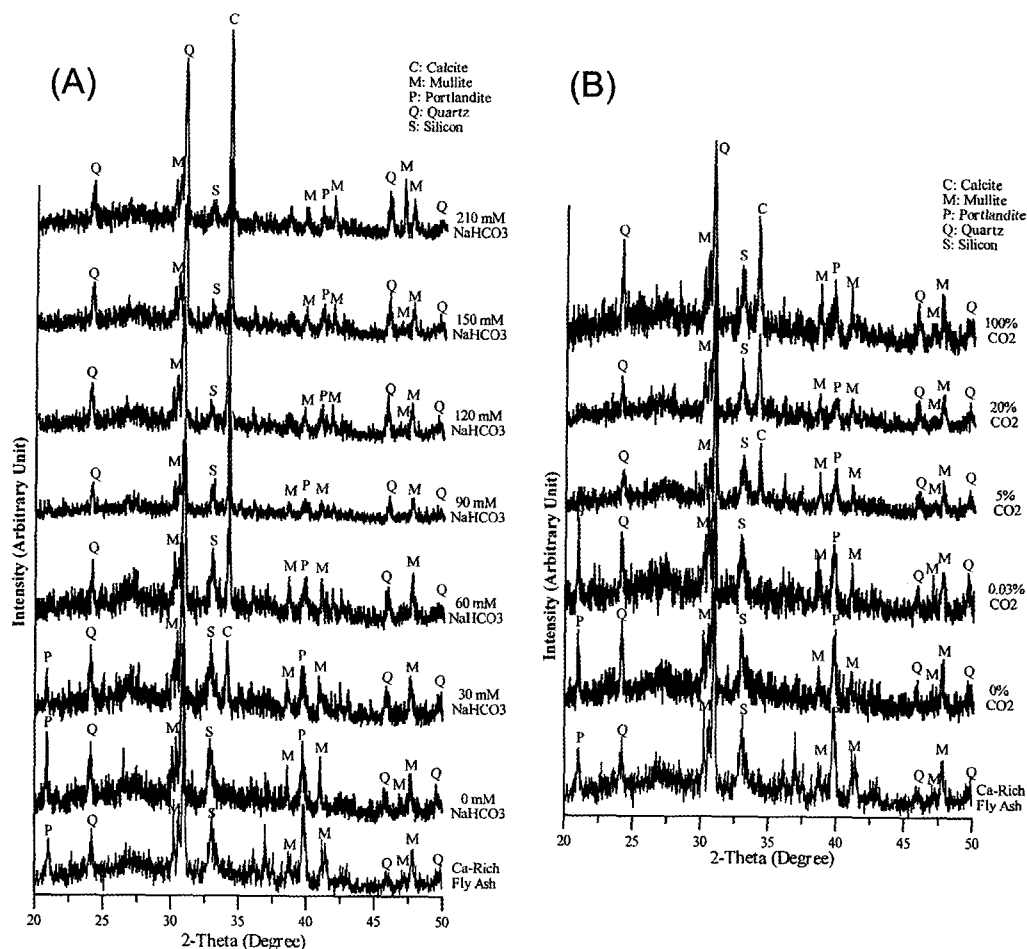


Fig. 2. Water soluble metals in the culture media after incubation using Ca-rich Springerville fly ash and MRB used for carbon sequestration (20% CO<sub>2</sub>).



**Fig. 3.** XRD analysis of Ca-rich fly ash used for carbon sequestration. (A) differences in NaHCO<sub>3</sub> concentration (0-210 mM), (B) differences in pCO<sub>2</sub> (0-100% CO<sub>2</sub>).

and Fe-rich fly ash. These upscaled experiments show potential for dramatic improvements in carbon and metal sequestration by complementing existing fly ash handling with biogeochemical processes. This study indicates that siderite and calcite precipitation using MRFA is generally associated with the bacterial metabolism of organic matter and hydrogen coupled with microbial Fe(III) reduction in the presence of reducing environments and high bicarbonate buffer or a H<sub>2</sub>/CO<sub>2</sub> atmosphere. High alkalinity and Fe(II) ions, as prompted by bacterial activity, seem important to biologically facilitated precipitation of carbonate minerals. The microbial production of Fe(II) and lowered redox potential

(Eh) also stimulates siderite precipitation (Fredrickson *et al.*, 1998; Zhang *et al.*, 1998; Roh *et al.*, 2003).

The MRFA, rejected dust from cement kilns and non-regulated agricultural wastes or food processing wastes are often trucked and land filled at total disposal costs (including transportation) often exceeding \$50 per ton while CO<sub>2</sub> is liberated to the atmosphere. While previous strategies dealt with these as separate issues, there may be an opportunity to energyplex them into useful product lines in high ionic strength ponds. The high ionic strength would be provided by the fly ash (along with residual sulfate from the sulfur in the coal) and/or reject kiln dust.



CO<sub>2</sub> from the plant could be bubbled through the deep alkaline pond. Agricultural wastes could provide additional energy into the pond. Anaerobic bacteria fed by the organics in the wastes precipitate additional carbonates to the sediments. In our laboratory different cultures of these bacteria produce siderite pellets in 5% salts at temperatures from 4 °C to 75 °C within days. Importantly, they also reduce other metals including chromium, manganese, uranium, and cobalt (Zhang *et al.*, 1996). The product of our proposed process could therefore be a hydrated multi-metal limestone-like aggregates suitable for road fill materials or other uses.

### Implications

Sequestration mechanisms would include biogeochemical processes and abiotic geochemical precipitation. To the mixture of ash, agricultural wastes, water and bubbled CO<sub>2</sub> one could add of tons of waste cement kiln dust per day enhancing the bio- and geo-chemical precipitation and sequestration efficiency. Accordingly, a fraction of carbon per cubic foot of pond each day (one pound per day per 10 cubic ft of pond) could be sequestered. Such an efficiency would represent approximately 10% of the efficiency of our microbial cultures observed in the laboratory in the absence of significant abiotic geochemical precipitation. By circulating warm process waters the biogeochemical sequestration rates would dramatically increase. Using the same conservative assumptions field sequestration may represent less than 1% of the metabolic efficiency of our lab cultures. Therefore, use of warm recirculating waters to heat the ponds could accelerate rates of sequestration and/or require less land for the biogeochemical sequestration.

### Conclusions

Given the abundance of Fe and Ca in MRFA, the capacity of Fe(III)-reducing bacteria to precipitate carbonate minerals could have a significant impact on carbon sequestration. In addition

to the precipitation of carbonate minerals via the biogeochemical processes, the microbial utilization of organic matter and hydrogen also contributes to (in)direct (co)precipitation of redox sensitive metals in fly ash ponds. The capacity of iron-reducing bacteria to precipitate carbonate minerals using fly ash creates the possibility of more effective CO<sub>2</sub> sequestration. In environments with high bicarbonate concentrations, the microbial production of Fe(II) from Fe-rich fly ash stimulates siderite formation. Biological carbonate mineral formation using fly ash indicates that biogeochemical processes may complement the capture of CO<sub>2</sub> from fossil fuel plants while potentially stabilizing fly ash wastes.

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