



Comparison of alkaline industrial wastes for aqueous mineral carbon sequestration through a parallel reactivity study



Clinton W. Noack^{a,*}, David A. Dzombak^a, David V. Nakles^a, Steven B. Hawthorne^b, Loreal V. Heebink^b, Neal Dando^c, Michael Gershenzon^c, Rajat S. Ghosh^c

^a Department of Civil and Environmental Engineering, Carnegie Mellon University, 5000 Forbes Avenue, Pittsburgh, PA 15213, United States

^b Energy and Environmental Research Center, University of North Dakota, 15 North 23rd Street, Stop 9018, Grand Forks, ND 58202, United States

^c Alcoa Technical Center, 100 Technical Drive, New Kensington, PA 15068, United States

ARTICLE INFO

Article history:

Received 5 December 2013

Accepted 18 March 2014

Available online 13 April 2014

Keywords:

Carbon sequestration

Alkaline industrial wastes

Mineral carbonation

Chemical equilibrium modeling

ABSTRACT

Thirty-one alkaline industrial wastes from a wide range of industrial processes were acquired and screened for application in an aqueous carbon sequestration process. The wastes were evaluated for their potential to leach polyvalent cations and base species. Following mixing with a simple sodium bicarbonate solution, chemistries of the aqueous and solid phases were analyzed. Experimental results indicated that the most reactive materials were capable of sequestering between 77% and 93% of the available carbon under experimental conditions in four hours. These materials – cement kiln dust, spray dryer absorber ash, and circulating dry scrubber ash – are thus good candidates for detailed, process-oriented studies. Chemical equilibrium modeling indicated that amorphous calcium carbonate is likely responsible for the observed sequestration. High variability and low reactive fractions render many other materials less attractive for further pursuit without considering preprocessing or activation techniques.

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1. Introduction

Precipitation of stable carbonate minerals provides a means for capturing and sequestering carbon dioxide (CO₂) in aqueous scrubber solutions. Alkaline industrial wastes are potential sources of polyvalent cations, especially calcium and magnesium, for reaction with aqueous carbonate and precipitation of carbonate minerals. In addition to the carbon sequestration that is achieved, this process provides an opportunity for beneficial reuse of abundant industrial wastes.

Interest in aqueous mineral carbonation as a large-scale option for carbon sequestration arose from observations of natural silicate weathering and abundance of thermodynamically suitable raw materials (Seifritz, 1990). Owing to their ubiquity, these minerals (chiefly magnesium silicates such as olivine and serpentine) continue to be the focus of research for myriad process schemes, treatment additives, and condition optimizations (Zevenhoven et al., 2011). Similarly, industrial waste materials with abundant alkaline contents such as fly ash, iron and steel slag, and alumina refining wastes have been studied as alternatives to mined ores in direct,

gas–solid carbonation schemes as well as aqueous carbonation schemes, the focus of this work (Bobicki et al., 2012).

Variability in the mineralogies of the cation-source materials along with the dynamic process conditions under which they are generated complicates comparison of results between independent studies. This issue is exacerbated by the wide variety of experimental designs and analytical tools used by researchers to investigate the carbon sequestration capacity of these materials. For example Montes-Hernandez et al. (2008) determined a carbon sequestration capacity of 26 g CO₂ per kg of fly ash in a batch system while Back et al. (2008) found a capacity of 230 g CO₂ per kg of fly ash in a flow-through design. Hence, it is useful and necessary to examine a variety of materials under identical conditions to gain insight into their relative behaviors.

Aqueous CO₂ capture technologies described in the literature commonly examine the use of the ubiquitous enzyme carbonic anhydrase (CA) to overcome slow CO₂ hydration kinetics (Bond et al., 2001; Favre et al., 2009; da Costa Ores et al., 2012; Vinoba et al., 2010). Studies that have examined CA systems typically used brines, either natural or synthetic, as the polyvalent cation source, and have inadequately assessed the potential role for alkaline industrial wastes (Liu et al., 2005; Favre et al., 2009; Rawlins, 2008). Alternatively, use of sodium hydroxide (NaOH) to capture CO₂ from ambient air has been proposed, with CO₂ sequestered

* Corresponding author. Tel.: +1 4436556602.

E-mail address: cnoack@andrew.cmu.edu (C.W. Noack).

via causticization and calcination prior to geologic storage (Zeman, 2007; Stolaroff et al., 2008). For this study, sodium bicarbonate is used to mimic the chemistry of a solution that might result from buffered CO₂ capture with CA or from NaOH absorption.

This work examined directly comparable reactivities of a wide range of alkaline industrial wastes by exposure to a simple sodium bicarbonate solution with a goal of providing guidance to future, process-oriented studies of these materials. The overall goal of the study was to investigate relative efficiencies of alkaline industrial waste reaction with aqueous carbonate. The specific objectives were to: estimate the relative potential aqueous carbon sequestration capacities of various alkaline industrial wastes; estimate the reaction time necessary to reach short-term pseudo-equilibrium; and determine the most promising high-volume industrial wastes for use in an aqueous carbon sequestration process.

2. Materials and methods

2.1. Industrial residuals

The types of materials investigated in this study were selected based on reported mineral compositions and related prior work. Thirty-one samples representing several distinct industrial operations were acquired (see Table 1). The test samples included: coal combustion fly ash (FA), spray dryer absorber ash (SDA), circulating dry scrubber ash (CDS), cement kiln dust (CKD), blast- or basic oxygen-furnace slag (FS), electric arc furnace dust (EAFD), and wet flue gas desulfurization gypsum (GYP). Additionally, two benchmark samples that were previously studied (Dilmore et al., 2009) – fly ash and spray drier absorber ash – were obtained from the National Energy Technology Laboratory (NETL) of the U.S. Department

Table 1

Matrix of samples acquired for study. Sample types are: coal combustion fly ash (FA), spray dryer absorber ash (SDA), circulating dry scrubber ash (CDS), cement kiln dust (CKD), blast- or basic oxygen-furnace slag (FS), electric arc furnace dust (EAFD), and wet flue gas desulfurization gypsum (GYP). Boiler and fuel types specified by sample providers.

Sample ID	Sample type	Boiler type	Fuel type
FA-1	FA	Pulverized coal (PC)	Lignite
FA-3	FA	Cyclone	Lignite
FA-5	FA	PC	Lignite
FA-4	FA	Cyclone	Lignite
FA-7	FA	Gassifier	Lignite
FA-8	FA	Spreader stoker	Lignite
FA-6	FA	Fluidized bed	Lignite
FA-11	FA	Cyclone	Subbituminous
FA-2	FA	PC	Subbituminous
FA-9	FA	PC	Subbituminous
FA-10	FA	PC	Subbituminous
FA-DOE	FA		
FA-12	FA	PC	S/B Blend
SDA-3	SDA	PC	Lignite
SDA-2	SDA	Cyclone	Lignite
SDA-5	SDA	PC	Subbituminous
SDA-7	SDA	PC	Subbituminous
SDA-1	SDA	PC	Subbituminous
SDA-4	SDA	PC	Subbituminous
SDA-DOE	SDA		
SDA-6	SDA		Bituminous
CDS-1	CDS	Industrial	
CDS-2	CDS		Bituminous
CKD-1	CKD	N/A	N/A
CKD-2	CKD	N/A	N/A
CKD-3	CKD	N/A	N/A
FS-1	BOFS	N/A	N/A
FS-2	BFS	N/A	N/A
EAFD-1	EAFD	N/A	N/A
EAFD-2	EAFD	N/A	N/A
GYP-1	GYP		

of Energy. All SDA and CDS samples came from coal-fired power plant operations without fly ash pre-collection and thus represent a blend of combustion fly ash and desulfurization solids. Samples FA-1, FA-2, FA-3, FA-5, FA-9, FA-10, SDA-1, SDA-2, SDA-3, and SDA-4 were part of the EERC coal combustion product (CCP) sample bank and had been analyzed as part of a previous study. No discernable, systematic differences were observed between the previously analyzed and freshly collected samples.

2.2. Preliminary solid composition characterization

The bulk chemical composition of all samples was determined using X-ray fluorescence (XRF) spectroscopy. Two XRF techniques (fused pellet and pressed pellet) were employed for the analysis of the samples (ASTM, 2004; ASTM, 2011). The EERC CCP sample bank samples were analyzed as fused pellets while all other samples were analyzed as pressed pellets.

2.3. Leaching methodology for screening and detailed experiments

2.3.1. Leaching protocol

Prior to testing, the samples were ground, if necessary, so that 80% of the sample mass passed through a 1 mm screen. Twenty grams of sample were mixed with 120 mL of 0.5 M NaHCO₃ solution at 350–500 RPM by a magnetic stir bar in a 500 mL glass beaker. The sodium bicarbonate solution was prepared using distilled water. Similar tests were conducted with distilled water (specific conductivity <20 μmhos/cm) as the extraction agent. Measurements of pH were taken of the stirred slurry throughout the experiment. pH meters were calibrated using standard buffer solutions at pH 7, pH 10, and pH 12.

After the contact period, the slurry was allowed to settle briefly. The supernatant was decanted, centrifuged, and then filtered at 0.45 μm. Finally, five grams of the filtrate was titrated to an endpoint of pH 4.5 (and an intermediate endpoint of pH 8.3) with 1.0 N HCl to determine alkalinity. Each filtered supernatant was titrated in duplicate.

2.3.2. Screening criteria

Screening tests were conducted on the entire suite of solids for 24 h in each leach solution with procedural duplicates in 0.5 M NaHCO₃. Samples were ranked according to reactivity and sequestration capacity. These screening tests were used to focus subsequent, detailed testing and analysis on the best-performing samples. Reactivity was assessed based on the time evolution of slurry pH while sequestration capacity was calculated using final pH and alkalinity (Section 2.6). Screening-stage leachates and residual solids received no additional analysis.

Based on reactivity and calculated carbon sequestration, the most reactive solids were selected for more detailed investigation. To identify the subset of samples for in-depth testing and modeling, consideration was given to identifying the most promising carbon sequestration candidates while also ensuring that samples from each of the material types were included, with highly similar samples being excluded to avoid redundancy.

2.3.3. Detailed leaching experiments

Leach testing on the set of best-performing samples was conducted in distilled water for four hours and in 0.5 M NaHCO₃ for one and four hours. As with the screening tests, pH was measured and the slurry was separated at the end of the test period with a filtered supernatant sample titrated to determine alkalinity. Additionally, supernatants and solids were reserved for further analysis with the solids being dried at 42–48 °C to preserve any hydrated minerals which may have precipitated.

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