



Experiments Summarizing the Potential of CO₂ Sequestration in the Basalts of Massachusetts – Final Report

Compiled by: Carrie Petrik and Stephen B. Mabee



Prepared for:

Massachusetts Clean Energy Center Boston, MA

October 15, 2011

Additional copies of this report and all accompanying documentation are available upon request by contacting the Massachusetts Geological Survey.

Address:

Massachusetts Geological Survey Department of Geosciences University of Massachusetts 611 North Pleasant Street Amherst, MA 01003 Telephone: 413-545-4814 Fax: 413-545-1200 Email: sbmabee@geo.umass.edu World Wide Web: http://www.geo.umass.edu/stategeologist/

Authors: Carrie Petrik, Department of Geosciences, University of Massachusetts at Amherst Stephen B. Mabee, State Geologist, University of Massachusetts at Amherst

Acknowledgments:

This work was supported by a grant (Task Order 09-1) from the Massachusetts Clean Energy Center to the University of Massachusetts. The authors wish to acknowledge the technical guidance and assistance of Drs. Steven Petsch, Sheila Seaman, David Finklestein, Mike Jercinovic and Chris Koteas at the University of Massachusetts at Amherst. Appreciation is also extended to Tom Naughton for help with grain size analyses, Pete Dawson for XRF analysis support, John Sweeny for experimental set-up assistance and Mike Vollinger for geochemical analysis assistance.

Cover: SEM image of carbonate minerals formed in Deerfield Basalt. The white mineral on bottom edge of gray grain is carbonate precipitated on plagioclase mineral after reaction with CO₂ at 1400 psi and 100°C for three months.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Executive Summary

Basalts are gaining more attention as reservoirs for the geological sequestration of carbon dioxide (CO_2). The purpose of this report is to present the results of experiments that were conducted on the basalts in western Massachusetts and Connecticut to determine their potential to sequester CO_2 . There were two primary objectives of these experiments:

- 1. To recreate and validate prior carbonate mineralization experiments conducted on the Holyoke basalt by Schaef et al. (2009) from Pacific Northwest National Laboratories (PNNL) and to test if their results are reproducible and geographically consistent within western Massachusetts and Connecticut, and,
- 2. To explore the possibility of reacting CO_2 with basalt at the earth's surface in an ex-situ mineral reactor and, in particular, to identify the optimum conditions necessary to precipitate large amounts of carbonate at the surface in a short time period by varying pressure, temperature, water volume, mass of sample and grain size in the experiments.

Sequestration of CO_2 in basalt is accomplished by mineral trapping where CO_2 , usually at supercritical conditions (>1000 psi and >31°C), mixes with water and reacts with the plagioclase and pyroxene minerals in the basalt to form stable carbonate minerals. Mineral trapping is the most secure way of sequestering CO_2 .

Basalts occur in the Connecticut River Valley of Massachusetts and Connecticut as a series of extrusive lava flows 300 to 400 feet thick and are exposed in the central part of the valley. For this study, three samples of basalt were collected and tested; the Holyoke basalt from the Tariffville Gorge near Granby CT, the Holyoke basalt from the Holyoke range in Amherst, MA and a sample of Deerfield basalt from the Cheapside Quarry in Greenfield, MA.

Seven experiments were conducted on powdered samples of the basalt. The powdered samples were placed in pressure vessels with varying amounts of deionized water and flooded with pure CO_2 . Pressure, temperature, volume of water, mass of sample, grains size and duration of test were varied in each experiment to evaluate the conditions necessary to produce various phases of carbonate.

Main conclusions from this work are as follows:

- 1. All three samples of basalt consistently produced carbonate. The Holyoke basalt produced more carbonate than the Deerfield basalt except when the experiments were run for at least 3 months. In that case, the Deerfield basalt produced more.
- 2. Carbonate mineralization can be produced under non-supercritical conditions (<1000 psi and <31°C).
- 3. Carbonate precipitation can occur rapidly, in as short as three hours, by reducing the grain size.
- 4. Each sample precipitates a unique set of carbonate minerals even if the whole rock chemistry is very similar. Thus, carbonate mineralization products and production rates are sensitive to minor shifts in chemical composition as well as geographic location.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011 5. The appearance of witherite ($BaCO_3$, barium carbonate) is an important catalyst for carbonate production. Once witherite appears, it precipitates quickly scavenging the carbonate ions from the CO_2 -water mix and increasing total carbonate production.

Optimum conditions for maximizing carbonate production include basalts with the following characteristics: 1) fine grain matrix; 2) >80 ppm barium content; and, 3) tholeiitic basalt composition (slightly alkaline conditions). Additional research is needed to create a more thorough list of characteristics.

Most research on sequestration in basalts has focused on deep injection of supercritical CO_2 into the basalt formation. A few researchers have explored the idea of an above ground, ex-situ mineralization process, using a pure mineral substrate to sequester CO_2 . The results of this work, however, have demonstrated favorable results from experiments that explore basalts as the feed stock for an ex-situ mineralization process, rather than a single mineral. In addition, these new experiments show that conversion of CO_2 to stable carbonate occurs quickly (within 3 hours) and under non-supercritical conditions, which has implications for either an ex-situ or in-situ application.

These results suggest also that public acceptance of carbon capture and storage might be more favorable through the use of basalts in an above ground batch reactor that is designed for individual, local scale carbon generators. There is no leakage hazard from the subsurface, no expensive pipelines required to transport supercritical CO_2 from the source to injection point, no extensive post injection monitoring needed, and no extensive reservoir modeling required including the inherent uncertainties attendant with any modeling exercise. The basalt residue from normal quarry or trap rock operations can be delivered to the individual power plant and then removed for reuse once reacted.

Challenges to an ex-situ carbon mineralization process remain. Additional steps needed to provide proof of concept include:

- 1. Energy and Life Cycle Analysis a full and complete energy audit needs to be conducted to evaluate economic feasibility.
- 2. Small-Scale Pilot Study The bench top experiments need to be scaled up using larger amounts of material and reactors. Will sequestration work if gases other than CO_2 are in the input stream and the gas is actively flowing through the batch reactor? In our experiments, the CO_2 was not flowing.
- 3. Optimization Study More basalts from the around the world need to be tested under varying experimental conditions to isolate those conditions leading to maximum carbonate production. Results appear to be sensitive to minor changes in chemistry.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Table of Contents

| Executive Summary | iii |
|--|------|
| Chapter One: Introduction | 1 |
| 1.1 Purpose of Work | 1 |
| 1.2 Objectives | 1 |
| 1.3 History of Project | 2 |
| 1.4 Background | 3 |
| Chapter Two: Geological Storage of CO ₂ in Basalt | 5 |
| 2.1 Basalts as Storage Reservoirs | 5 |
| 2.2 Basalts of the Connecticut Valley | 7 |
| 2.3 Previous Work on Basalts of the Connecticut Valley | . 13 |
| Chapter Three: Experimental Set-Up and Methods | . 15 |
| 3.1 Sample Preparation | . 17 |
| 3.2 Experimental Design | . 18 |
| 3.3 Experimental Description | . 19 |
| 3.4 Experimental Apparatus and Materials | . 22 |
| 3.5 Methods | . 24 |
| Chapter Four: Results | . 27 |
| 4.1 Rock Chemistry and Mineralogy | . 27 |
| 4.2 Carbonate Production | . 27 |
| 4.3 Summary of Results | . 34 |
| Chapter Five: Summary and Conclusions | . 38 |
| Chapter Six: Implication of Results and Next Steps | . 39 |
| 6.1 Next Steps | . 40 |
| 6.2 Carbonate Mineralization and the State of Massachusetts | . 41 |
| References Cited | . 43 |
| Appendices | . 45 |
| Appendix A: X-Ray Diffraction (XRD) Data | . 45 |
| Appendix B: X-Ray Fluorescence (XRF) Data | . 55 |
| Appendix C: SEM Spectra and Images | . 56 |
| Appendix D: SEM Elemental Mapping | . 86 |
| Appendix E: Grain Size Data | . 93 |

Chapter One: Introduction

1.1 Purpose of Work

The following is the final report for the Massachusetts Clean Energy Center to provide an assessment of the feasibility of sequestering carbon dioxide in local basalt formations. Carbon sequestration is the permanent storage of carbon dioxide gas [CO₂] for the purpose of mitigating accumulated CO₂ in the atmosphere. Carbon sequestration is part of the proposed technique known as carbon capture and storage [CCS], in which CO₂ is separated from other gases and combustion products at a point source, such as a fossil fuel-based power plant, transported to a storage location, and injected into deep geological formations (IPCC, 2005). Carbon sequestration can be accomplished through a variety of approaches. These include biological/terrestrial storage, injection into deep ocean waters and sediments, and permanent storage in geologic reservoirs. Biological storage relies on removal of atmospheric CO₂by plant biomass or soil carbon. Ocean storage relies on high pressure found at great depths to convert CO₂ into a supercritical liquid. Geological sequestration involves pumping CO₂ into deep underground reservoirs of rock that have the capacity to permanently take up CO₂. Geological units that are considered appropriate for the geological sequestration of CO₂ are deep saline aquifers, depleted oil and gas fields, unmineable coal seams and basalts. When carbon dioxide is stored in basalts a geochemical process occurs that results in the precipitation of carbonate minerals. This process of carbonate mineralization permanently locks the carbon dioxide gas into a solid mineral. The purpose of this report is to outline findings from experiments that were conducted on the basalts of western Massachusetts and Connecticut to determine their potential to geologically sequester CO₂.

1.2 Objectives

The work presented here was designed around two main objectives. The first objective was to recreate and validate prior carbonate mineralization experiments conducted on the Holyoke basalt by Schaef et al., (2009) from Pacific Northwest National Laboratories (PNNL). The work from PNNL sampled the Holyoke basalt in its most southern location in south central Connecticut as well as collected samples from around the world. In the PNNL study the Holyoke basalt performed very well and was found to precipitate carbonate the fastest of all of the samples taken. Therefore, we wanted to examine if the findings of PNNL were applicable to our local Holyoke and neighboring Deerfield basalts.

The second objective of our work was to examine the basalts of the Connecticut River Valley for their potential to sequester carbon dioxide. Compared to current CCS projects the Connecticut River Valley as a basin, in terms of its areal extent, is quite small. This, however, does not mean that the region is without promising options in terms of geologically sequestering carbon dioxide. One option for the region is to examine the potential of the local basalts to sequester CO_2 in a traditional situation by pumping CO₂ underground. To do this would require an extensive drilling operation into the geology to determine the geochemical profile of the basalt as well as the structural geology and hydrology of the area. The second option is to explore the possibility of reacting CO₂ with the basalt at the surface in an ex-situ mineral reactor. In this situation an

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

expensive drilling program is not necessary. Therefore, the main focus of the second objective is to determine what conditions, in terms of pressure, temperature, water volume, mass of sample and grain size are necessary to precipitate large amounts of carbonate in a short time period.

1.3 History of Project

This report on sequestration of CO_2 in basalts is part of a larger project under Task Order 09-1 that examined the feasibility of carbon storage in five geologic repositories across Massachusetts. These repositories included deep saline aquifers in the Connecticut River Valley, unmineable coal beds in the Narragansett basin, organic-rich shales in the Connecticut River Valley, deep basalts in the Connecticut River Valley and organic-rich schist and slate in the Berkshires (Figure 1.1). The results of this larger study are articulated in a series of four reports that were submitted previously to CEC.



Figure 1.1. Map showing general areas (gray areas) where potential targets for geologic carbon sequestration in Massachusetts might be possible. These include: A) deep saline aquifers in the New Haven arkose and Sugarloaf arkose in the Connecticut Valley; B) Organic rich shales in the Connecticut Valley; C) unmineable coal beds in the Narragansett basin; D) deep basalts in the Connecticut Valley; and, E) carbon-bearing graphitic schists in the Berkshires.

The first report was submitted in February 2010. This report summarized the results of a data mining exercise to determine what information is known about each potential repository in Massachusetts and what additional data are necessary and must be attained from analogous basins.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

The second report, submitted in May 2010, provided a preliminary assessment of the CO_2 storage resource potential in two of the five target areas in Massachusetts. These included the deep saline aquifers of the Connecticut Valley and the unmineable coal beds of the Narragansett Basin in southeastern Massachusetts. Where data mining and new analyses provided data, these were directly input into the carbon storage calculation models. Where information was uncertain or lacking, range estimates complete with full error analysis were applied. The organic-rich shales and schists/slates of the Connecticut Valley and Berkshires, respectively, were not considered further as targets due to their limited extent and lack of a suitable model from the National Energy Technology Laboratory to estimate storage potential. Storage in basalts was deferred for later analysis.

The third report was identical to the second report except that a chapter on the implications of results was added to help identify next steps in the study, determine if further research on CO₂ storage in Massachusetts is justified and identify approximate costs for additional research. This report was submitted in June 2010.

One of the outcomes of the third report was the need for the collection of adsorption data on Narragansett coals to obtain a better estimate of storage potential. During the summer of 2010, additional samples of coal from the Narragansett basin were obtained and analyzed by an outside laboratory to determine actual adsorption capacity. This new data was incorporated in a fourth report that was submitted on May 15, 2011.

Another outcome of the third report was the need to evaluate further the potential of basalt to sequester CO₂ through a chemical reaction that produces a harmless carbonate precipitate. Previous work by Schaef et al. (2009) suggested that Massachusetts basalts were able to sequester carbonate effectively. We believed this warranted further examination and with a second no-cost extension granted by CEC, we embarked on a series of bench top experiments to evaluate more fully the potential of sequestering CO_2 in basalt.

The present report, due October 15, 2011, represents the fifth and final report in this larger Task Order. It summarizes the results of a set of experiments and analyses conducted over a 1 year period that examine in detail the propensity of Massachusetts basalts to convert CO₂ into carbonate minerals.

1.4 Background

Basalts are gaining more attention as reservoirs for the geological sequestration of carbon dioxide (McGrail et al., 2006; Alfredsson et al., 2008; Charan and Begum, 2008; Goldberg et al., 2008; Oelkers et al., 2008). Alfredsson et al., (2008) describe the effects of injecting CO₂ rich waters into basalts in southwest Iceland. They concluded that the basalt formations would release large amounts of cations that would result in the formation of carbonate minerals. Reaction path modeling has been used by others to predict the formation of calcite and dolomite in Icelandic basalts when exposed to CO₂ rich waters (Gysi and Stefansson, 2008; Flaathen et al., 2009). This concept will be tested with the injection of 30,000 tons of CO₂ per year into basalt formations at Hellisheidi, Iceland (Oelkers et al., 2008). This pilot project is being led by a team out of Columbia University. To date, the team has completed the drilling of wells, surface monitoring

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

for baseline data and hydrologic studies. In 2010 a carbon capture unit was attached to a geothermal power plant. Injection is expected by the end of 2011. Laboratory results were reported also from researchers in India about the possibility of the Deccan basalts being utilized for carbon sequestration (Charan and Begum, 2008). On the Deccan basalts, it was found that the predominant carbonate mineral precipitated was ankerite. A similar report was also conducted in Washington State on the Columbia River basalts by McGrail et al. (2006). This work investigated the kinetics of basalt water interactions and found that carbonate precipitation happens within a time frame that makes the sequestration of CO_2 in basalts a viable option. The Big Sky Regional Carbon Sequestration Partnership recently conducted a small-scale pilot study into the Columbia River basalt in Washington State to investigate the mineralogical, geochemical and hydrologic impact of injected CO_2 on the basalt formation. This is the first basalt field test in the nation. Thus far, wells have been drilled, but injection has yet to begin.

When considering a basalt formation as a possible storage reservoir for geological sequestration similar criteria are used as for deep saline aquifers.

- 1. Adequate capacity to hold the proposed amount of CO₂
- 2. Ability to pump fluid into the formation at an established rate and pressure without damaging (fracturing) the rock (injectivity)
- 3. Presence of a caprock directly above the unit which would aid in restricting movement of CO₂ in the vertical direction
- 4. A geologic environment that lacks features that would render it unstable, such as faults or hydrothermal activity.

As with deep saline aquifers, a basalt CCS project includes the capture, compression, transport, and injection of carbon dioxide. Basalts, however, have a unique mineralogy which, when combined with CO_2 rich water, react to precipitate carbonate minerals. This reaction essentially locks the CO_2 gas permanently into a carbonate mineral.

Chapter Two: Geological Storage of CO2 in Basalt

2.1 Basalts as Storage Reservoirs

Basalt is a geologic formation that is the result of solidified lava flows at earth's surface as well as injection of lava as horizontal sheets and vertical intrusions. Individual flows and intrusions typically are only a few meters thick, but repeated volcanic eruption and magmatic activity can build up thick sequences of basalt that can be 10s to 100s of meters thick or greater. Basalt is made of mafic minerals, that is, minerals that are rich in iron, magnesium and calcium such as anorthite, (calcium-rich plagioclase feldspar), pyroxene, amphibole and olivine. These minerals can react with CO_2 under elevated pressures and temperatures, consuming the primary minerals and generating new carbonate minerals, such as calcite. Naturally-occurring reactions between basalt and dissolved CO_2 develop along mid-ocean ridges in seafloor hydrothermal systems. Artificial reactions of basalt minerals with CO_2 have also been demonstrated (McGrail et al., 2006), thus leading to basalts being proposed as potential permanent CO_2 storage reservoirs.

Various types of large basalt flows exist through the world and U.S. (Figure 2.1). Large igneous provinces (LIPs) represent immense outpourings of mafic magma and include continental flood basalts (CFBs), passive volcanic margins, sedimentary rift basins and oceanic plateaus. Because of the large geographical distribution of basalt flows it seems that these geological units have some of the necessary characteristics of storage capacity, extent and permeability to support geological sequestration of CO₂.



Figure 2.1. Map showing basalts of the United States in relation to major CO_2 sources. Photo courtesy of Idaho National Labs.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Each type of geological reservoir (saline aquifer, depleted oil and gas fields, unmineable coal seam, etc.) has specific trapping mechanisms associated with it (Figure 2.2). When carbon dioxide and water are injected into a basalt formation the main trapping mechanism is mineral trapping via mineral carbonation. Mineral trapping is considered to be the most secure form of carbon storage because once the chemical reactions occur the carbon dioxide is now permanently stored as a solid mineral.



Figure 2.2. Illustration of various types of trapping mechanisms over time and associated storage security. Trapping means the various mechanisms that keep the carbon dioxide, once underground, in place. When CO_2 is initially injected into a reservoir the CO_2 is kept within that reservoir because of the geological caprock above the storage unit. This is referred to as stratigraphic trapping. Over time, different trapping mechanisms become more active in holding the CO_2 within the reservoir.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Mineral trapping occurs through a series of water rock chemical reactions. CO_2 solubility decreases with an increase in temperature and ionic concentration of formation waters and increases with pressure. The dissolution of CO_2 acidifies formation waters through the following reactions.

 $CO_2(aq) + H_2O = H_2CO_3$ (carbonic acid) $H_2CO_3 = HCO_3$ -(bicarbonate) + H⁺ $HCO_3 = CO_3^{2-}(carbonate) + 2H^+$ (1)

First, aqueous CO_2 reacts with water to produce carbonic acid. Next, the carbonic acid will disassociate into bicarbonate ions and carbonate ions (reaction 1). If divalent cations (such as Ca, Mg, or Fe) are in solution, carbonate precipitation will occur.

$$(Ca,Mg,Fe)^{2+} + HCO_3^- = (Ca,Mg,Fe)CO_3(solid) + H^+$$
 (2)
 $(Ca,Mg,Fe)^{2+} + CO_3^{2-} = (Ca,Mg,Fe)CO_3(solid)$ (3)

Unless the H+ ions produced during reactions 1 and 2 are consumed, no further reactions can occur. Calcium plagioclase dissolution (reaction 4) consumes H+ ions driving reactions 1 and 2 to the right resulting in carbonate precipitation (Matter and Kelemen, 2009).

$$CaAl_2Si_2O_8(plagioclase) + 2H^+ + H_2O = Ca_2^+ + Al_2Si_2O_5(OH)_4(clay)$$
 (4)

Therefore, in addition to temperature, pressure and salinity the consumption of CO_2 in formation waters is also controlled by the buffering of pH through fluid-rock interactions, which is controlled by the mineralogy of the host rock.

2.2 Basalts of the Connecticut Valley

There are two sedimentary basins located in the Connecticut River Valley in western Massachusetts that contain basalt (Figure 2.3). The northernmost basin is the Deerfield Basin and the southernmost basin is referred to as the Hartford Basin. Both are part of a larger chain of exposed sedimentary basins that extend along the eastern margin of North America from Nova Scotia to North Carolina, with subsurface basins extending further north into Newfoundland, south into Florida and the Gulf of Mexico, and seaward to the edge of the North American continental shelf (Figure 2.4). These basins formed as a result of crustal extension when the North American continent separated from the African continent to form the present-day Atlantic Ocean during the Middle- to Late Triassic, about 250 million years ago (Froelich and Robinson, 1988).

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.3. Map outlining the location of the Holyoke and Deerfield basalts within the Connecticut River Valley.

The Deerfield Basin trends north-south and averages 5 km (3 miles) wide by 25 km (15 miles) long (Taylor, 1991). The Deerfield Basin comprises five formations (Figures 2.4, 2.5, 2.6, 2.7). The oldest and deepest formation is the Sugarloaf Arkose. This formation is composed of fluvial sandstones (also known as red beds due to their red coloration from abundant iron oxidation) and is between 1600 meters (5280 feet) and 2370 meters (7800 feet) thick. Above the Sugarloaf Arkose are the Fall River Beds. These beds are 3 to 9 meters (10 to 30 feet) thick and are

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.4. Image showing the eastern coast of the Unites States indicates the location of continental rift basins along the eastern seaboard (shaded in black). Both the Deerfield and the Hartford basins are a part of this group and are shown in detail on the right. The Deerfield Basin is to the north while the Hartford Basin lies to the south and is in both Massachusetts and Connecticut.

black organic-rich shales. The Fall River Beds are overlain by the 120 meter (394-foot) thick Deerfield Basalt. This is a lava flow that flowed over the surface of the basin from rifts and vents

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.5. Map showing the distribution of the geologic units in the Deerfield Basin and the Hartford Basin in Massachusetts. The dividing line between the two basins is approximately along the line connecting Northampton and Amherst.

created during extension of the basin. The basalt consists of several flows that contain vesicles and openings in the upper part of the flow and individual flows at the surface often show columnar jointing. These are vertical joints that exhibit a characteristic hexagonal pattern on the surface and form as a result of cooling. The joints provide an excellent vertical pathway for fluids. The basalt is overlain by the Turners Falls formation that is between 90-1300 meters (295-4265 feet) thick. The Turners Falls formation consists of interbedded fine-grained sandstones and shales. The youngest formation in the Deerfield Basin is the 300-1000 meter (984-3280 foot) thick Mt. Toby Conglomerate. Although this unit is slightly younger than the Turners Falls Formation it is interbedded with the Turners Falls Formation.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.6. Generalized cross section of the Deerfield basin showing the juxtaposition of units in the subsurface. The Eastern Border Fault controlled the formation of the basin but numerous smaller faults are observed in surface exposures, especially in the Deerfield Basalt and adjacent strata. Trsa – Sugarloaf formation; Jfr – Fall River beds; Jdb – Deerfield basalt; Jmt – Mount Toby conglomerate; Jt – Turners Falls formation. From Walsh (2008).

The Hartford Basin is contiguous with the Deerfield Basin in central western Massachusetts. Most of this basin is in Connecticut, but the northern portion (some 25 km (15miles) wide and 30 km (19 miles) north-south) occupies the southern portion of the Connecticut River Valley in Massachusetts (Figures 2.4 and 2.7). The Hartford Basin exhibits a more complicated stratigraphy than the Deerfield Basin, with a 1830-meter (6000 foot) thick New Haven Arkose at its base (roughly equivalent to the Sugarloaf Arkose in the Deerfield Basin), three basalt layers (Talcott, Holyoke and Hampden basalts, each approximately 91 meters (300 feet) thick) interbedded with two formations consisting of shales and sandstones (the Shuttle Meadow and East Berlin Formations), topped with the approximately 2435-meter (8000foot) thick Portland Formation containing sandstone, shales and conglomerate.

Although initially deposited as horizontal, flat-lying strata of sediments, tectonic activity along the eastern border fault of the Deerfield and Hartford Basins approximately 200 million years ago caused these strata to dip to the east. The oldest strata (Sugarloaf Arkose in the Deerfield Basin and New Haven Arkose in the Hartford Basin) dip most steeply (up to 20° but can be as steep as 45° near the east margin of the basin), while the youngest strata (uppermost Mount Toby and Portland formations) are nearly flat-lying. For this reason, outcrops of the oldest strata along the western sides of these basins extend downward and eastward towards the eastern side of the basin.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.7. Stratigraphic columns of the Deerfield, Hartford, and Pomperaug Basins. (From Walsh, 2008) and complied from references therein.

The Deerfield basalt is about 120 meters (394 feet) thick and each of the basalts in the Hartford Basin average about 91 meters (300 feet) thick. The Deerfield basalt is pillowed (indicating it flowed into water) and vesicular (full of voids and openings) at its base whereas the upper portion of the basalt flow is strongly jointed. The basalts outcrop mostly in the center of the basin and form ridges due to their resistance to weathering (Figures 2.3 and 2.8). Approximately 52 km^2 (20 sq. miles) of basalt is exposed in the two basins. The basalts also dip to the east and south at 10° to 15° degrees (Figure 2.6).

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.8. Picture of Holyoke basalt outcrop from atop Mt. Holyoke.

2.3 Previous Work on Basalts of the Connecticut Valley

Previous work has explored the viability of volcanic flood basalts as a geologic storage option for CO₂ (McGrail et al., 2006; Alfredsson et al., 2008; Goldberg et al., 2008; Oelkers et al., 2008; Schaef et al., 2008; Schaef et al., 2009). McGrail et al. (2006) conducted an evaluation of the potential for the Columbia River Basalts in Washington State to be permanent geologic storage units. This laboratory study found that CO₂ saturated pore water reacted rapidly with the basalts to form stable carbonate minerals (McGrail et al., 2006). In 2009, Schaef et al. expanded on this work by experimenting on basalts from the eastern U.S., India and Africa. It was established that all basalt samples had similar bulk chemistry and common mineral assemblages as well as similar dissolution kinetics (Schaef et al., 2009). However, regardless of these similarities, long-term static experiments showed very different mineralization rates and precipitate chemistry.

Illustrated in Figure 2.9 are the results of the four basalt samples tested by Schaef et al, (2009). It is interpreted that a decrease in pressure observed in the Holyoke basalt (sample labeled Newark Basin) is a result of CO_2 reacting to form carbonate. Of the four basalts tested, the Holyoke Basalt showed the greatest variations in pressure. Once the experiment was concluded, the Holyoke Basalt also showed the most visible change of all the samples (Figure 2.10).

The preliminary results from Schaef et al., (2009) were promising. As a result, additional work was warranted to examine the propensity of carbonate formation in Massachusetts basalt under varying experimental conditions.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 2.9. Changes in carbon dioxide pressure, in psi, as a function of time, in days. Drops in pressure are interpreted to be due to carbonate mineralization. In this experiment the Newark Basin sample refers to a sample of the Holyoke basalt collected from the Hartford basin. Modified from Schaef et al., 2009.



Figure 2.10. Optical images of post-reacted Holyoke Basalt grains after 230 days of testing (Schaef et al., 2009).

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Chapter Three: Experimental Set-Up and Methods

Basalt samples were collected from three different locations, two in Massachusetts and one in Connecticut (Figure 3.1). Massachusetts samples were collected from the Cheapside Quarry in Deerfield and at the Notch in the Holyoke Range in Amherst Massachusetts (Figure 3.2). All samples were collected in August of 2010. At each sample location between 5 to 10 pounds of sample was collected. The Connecticut sample was taken from the Tariffville Gorge on the Farmington River just to the south east of Granby Connecticut (Figure 3.3).



Figure 3.1. Map showing the location where all three samples were collected.

Once collected, samples were crushed with a mortar and pestle to various grain sizes. Samples were then placed in a 25 ml Parr pressure vessel with deionized (DI) water. CO_2 gas was then heated to initiate the phase change to a supercritical fluid and was then injected into the pressure vessels. Temperatures and pressures were held constant for a specified amount of time. Once the experiment was completed the temperature was decreased and the pressure released from each vessel. The contents of each vessel was then removed and placed into a drying oven, at 60°C for 24 hours where all of the water was evaporated. Once dry, samples underwent various analytical techniques to quantify the geochemical changes that had occurred.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 3.2. Holyoke Basalt at the Notch off of Route 116 in Amherst, MA.



Figure 3.3. Image of the Tariffville Gorge on the Farmington River in northwestern Connecticut. Outcrops on either side of the river are Holyoke Basalt.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

3.1 Sample Preparation

Samples were initially broken to manageable pieces, roughly 3-inch chunks, using a hand held mallet. From this point they were placed in a mortar and pestle for further crushing where, again, a mallet was used to pound the pestle. Once pea sized grains were attained two different processes were followed, the samples were either placed in a sieve (.42 -2 mm) or into the shatter box.

One of the experimental parameters being examined in this work was grain size. In previous work by Schaef et al. (2009), they conducted experiments using a grain size distribution ranging from 0.42 mm to 2 mm. To replicate the work of Schaef et al. (2009), the pea size grains were sieved and the 2 mm to 0.42 mm fractions retained for analysis.

In addition, we also wanted to examine how carbonate mineral production is affected by reduced grain size. Accordingly, a portion of the pea sized grains from each sample was taken and placed in a Spew shatter box, which is a circular container of two concentric circles made of tungsten carbide that is clamped down within a wooded box. The tungsten carbide crushes the pea sized grains into a fine powder. Generally, the shatter box runs for about five minutes. The powder is then removed from the tungsten carbide cartridges and examined for grain size distribution to attain the average grain size (Figures 3.4 and 3.5).



Figure 3.4. Cumulative grain size distribution graph for the Deerfield basalt (experiments 1-6). The Y-axis, % passing, refers to the percentage of grains that passed through a given sized sieve. At 50% passing, half of the grains had fallen through the sieve. Therefore, the grain size at 50% passing is the median grain size. In this case the median grain size is 0.5 mm.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011





Figure 3.5. Cumulative grain distribution curves for Holyoke basalt in Connecticut (upper) and Massachusetts (lower). The median grain size is less than 0.5mm.

Thin sections of each sample were made also and examined under a microscope. Visual observations showed that both samples of the Holyoke Basalt were much finer grained than the Deerfield Basalt. The Deerfield Basalt had large angular class while the Holyoke Basalts exhibited a much finer, uniform texture. In all three samples the most predominate minerals were plagioclase feldspar and pyroxene.

3.2 Experimental Design

The goal of the experiments was to determine what conditions would promote the most abundant carbonate mineralization in order to create optimal sequestration. Conditions examined were pressure, temperature, water, grain size, mass and duration. The time needed to promote

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

carbonate mineralization was of particular interest in this work. Specifically, we wanted to see if by using smaller grain sizes it was possible to increase the amount of carbonate produced. To date, most work looking at carbon sequestration through carbonate mineralization, has only looked at reactions that involve CO_2 in a supercritical phase. Associated costs with the compression of CO_2 from a gas to a supercritical fluid are very high (Zero Emissions Platform, 2011). Therefore, we wanted to explore whether carbonate mineralization can be achieved effectively under non-supercritical conditions.

| Carbonate Species | Chemical Formula | |
|--------------------------|---------------------------|--|
| 01: | 0.00 | |
| Calcite | | |
| Dolomite | $(Ca,Mg)(CO_3)_2$ | |
| Ankerite | $Ca(Fe, Mg, Mn) (CO_3)_2$ | |
| Aragonite | CaCO ₃ | |
| Siderite | FeCO ₃ | |
| Strontianite | SrCO ₃ | |
| Witherite | BaCO ₃ | |
| Vaterite | CaCO ₃ | |

Figure 3.6. A list of all carbonate species investigated in this work.

Carbon dioxide, as with water, has different physical phases in which it can exist; solid, liquid, gas, and the less commonly known, supercritical phase (Figure 3.7). Pressure and temperature determine what phase will result. Low pressures and high temperatures, for example, will result in CO_2 being a gas whereas high pressures and low temperatures will result in a solid phase. The liquid phase occurs at mid temperatures and pressures. However, when both pressures and temperatures are high a supercritical phase occurs. As a supercritical fluid, the CO_2 adopts properties of both the gas phase and the liquid phase. Supercritical conditions exist above temperatures of $31.1^{\circ}C$ (88.3°F) and pressures of 7.39 MPa (1071 psi). Under these conditions the CO_2 will fill a container like a gas but with a density like that of a liquid.

3.3 Experimental Description

The following is a description of each experiment and the major research questions being examined in each experiment. For a quick reference of the experimental conditions of each experiment refer to Figure 3.8.

Experiment 1

This experiment was conducted above supercritical conditions (1400 psi and 100°C) and used the same water to sample ratio (4.8 ml water to 10 g sample) as was used in the original work of Schaef et al. (2009). The major departure in this experiment from the Schaef et al. (2009) work is

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 3.7. Phase diagram for carbon dioxide showing the different temperatures and pressures necessary for each phase to exist.

that this experiment only lasted for three hours and used a grain size of less than 0.5 mm. This departure from grain size and duration was maintained throughout most of the experimental work. The purpose of this experiment was to see if we could mineralize carbonate in a short time period (3 hours) with a reduced grain size. The conditions of experiment 1 are the baseline conditions used for all subsequent experiments. The only difference between experiment 1 and all others is that one of the main variables was altered (eg., pressure, temperature, water content, etc.).

Experiment 2

In experiment 2 all conditions are identical to experiment 1 except for pressure. In this experiment the pressure was reduced to 800 psi which resulted is the CO_2 being in a gas phase, rather than a supercritical phase. Therefore, the major research question in this experiment was will mineralization occur if CO_2 is in a gas phase?

Experiment 3

Again, baseline conditions from experiment 1 were used, except the temperature was reduced to 20° C from 100° C. In this experiment temperatures were low and pressures were high which

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| Experiment # | Deerfield Basalt (DB) | Holyoke Basalt Mass (HBM) | Holyoke Basalt Ct (HBC) |
|--------------|-------------------------|---------------------------|-------------------------|
| | Experimental Conditions | Experimental Conditions | Experimental Conditions |
| 1 | Time: 3 hr | Time: 3 hr | Time: 3 hr |
| | Pressure: 1400 psi | Pressure: 1400 psi | Pressure: 1400 psi |
| | Temp: 100°C | Temp: 100°C | Temp: 100°C |
| | Mass: 10g | Mass: 10g | Mass: 10g |
| | Water: 4.8 ml | Water: 4.8 ml | Water: 4.8 ml |
| | Grain Size: 0.5 mm | Grain Size: 0.5 mm | Grain Size0.5 mm |
| 2 | Time: 3 hr | Time: 3 hr | Time: 3 hr |
| | Pressure: 800 psi | Pressure: 800 psi | Pressure: 800 psi |
| | Temp: 100°C | Temp: 100°C | Temp: 100°C |
| | Mass: 10g | Mass: 10g | Mass: 10g |
| | Water 4.8 ml | Water 4.8 ml | Water 4.8 ml |
| | Grain Size: 0.5 mm | Grain Size: 0.5 mm | Grain Size: 0.5 mm |
| 3 | Time: 3 hr | Time: 3 hr | Time: 3 hr |
| | Pressure 1400 psi | Pressure 1400 psi | Pressure 1400 psi |
| | Temp: 20°C | Temp: 20°C | Temp: 20°C |
| | Mass: 10g | Mass: 10g | Mass: 10g |
| | Water: 4.8 ml | Water: 4.8 ml | Water: 4.8 ml |
| | Grain Size: 0.5 mm | Grain Size: 0.5 mm | Grain Size: 0.5 mm |
| 4 | Time: 3hr | Time: 3hr | Time: 3hr |
| | Pressure: 800 psi | Pressure: 800 psi | Pressure: 800 psi |
| | Temp 20°C | Temp 20°C | Temp 20°C |
| | Mass: 10g | Mass: 10g | Mass: 10g |
| | Water: 4.8 ml | Water: 4.8 ml | Water: 4.8 ml |
| | Grain Size: 0.5 mm | Grain Size: 0.5 mm | Grain Size: 0.5 mm |
| 5 | Time: 3 hr | Time: 3 hr | Time: 3 hr |
| | Pressure: 1400 psi | Pressure: 1400 psi | Pressure: 1400 psi |
| | Temp: 100°C | Temp: 100°C | Temp: 100°C |
| | Mass: 15g | Mass: 15g | Mass: 15g |
| | Water: 7.2 ml | Water: 7.2 ml | Water: 7.2 ml |
| | Grain Size: 0.5 mm | Grain Size: 0.5 mm | Grain Size: 0.5 mm |
| 6 | Time: 3hr | Time: 3hr | Time: 3hr |
| | Pressure:1400 psi | Pressure:1400 psi | Pressure: 1400 psi |
| | Temp: 100°C | Temp: 100°C | Temp: 100°C |
| | Mass: 10g | Mass: 10g | Mass: 10g |
| | Water: 10 ml | Water: 10 ml | Water: 10 ml |
| | Grain Size: 0.5 mm | Grain Size: 0.5 mm | Grain Size: 0.5 mm |
| 7 | Time: 3 month | Time: 3 month | Time: 3 month |
| | Pressure:1400 psi | Pressure:1400 psi | Pressure:1400 psi |
| | Temp100°C | Temp100°C | Temp100°C |
| | Mass: 25g | Mass: 25g | Mass: 25g |
| | Water: 12 ml | Water: 12 ml | Water: 12 ml |
| | Grain Size: .42- 2 mm | Grain Size: 42- 2 mm | Grain Size: 42- 2 mm |

Figure 3.8. Table summarizing the exact conditions of all experiments performed.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

resulted in CO_2 being in the liquid phase. Thus, we wanted to see if the liquid phase would produce carbonate mineralization.

Experiment 4

In experiment 4 both pressure and temperature were reduced, to 800 psi and 20°C respectively. All other parameters were identical to experiment 1. Again, this reduction of pressure and temperature resulted in CO_2 being in a gas phase rather than a non-supercritical state, similar to Experiment 2. However, this time both pressure and temperature were low. Therefore, questions were asked concerning if experiments 2 and 4 would produce similar results or does changing the pressure and temperature independently affect carbonate production?

Experiment 5

In experiment 5, CO_2 was again at a supercritical phase. All conditions were identical to experiment 1 except that the volume of sample and water was increased by 50% resulting in a water to sample ratio of 7.2 ml to 15 g. Here, we examined whether there was a linear relationship in the amount of carbonate produced between experiments 1 and 5.

Experiment 6

Experiment 6 examined the effect an increase of water had on the carbonate production of each sample. All conditions were the same as in Experiment 1 except, as in experiment 5, the water to sample ratio was changed. In this experiment the water to sample ratio was increased to 10 ml of water to 10 g of sample resulting in a 1/1 relationship. Does an increase in water, and therefore carbonic acid, change overall carbonate production?

Experiment 7

Experiment 7 is our attempt to recreate the Schaef et al. (2009) work using identical conditions, except for duration. Their work lasted upwards of 800 days, where ours, due to time constraints, only lasted 90 days. This experiment was at supercritical conditions using 25 grams of sample, 12 ml of water and a grain size range of 0.42-2 mm.

3.4 Experimental Apparatus and Materials

All experiments were conducted using a 25 ml Parr pressure vessel constructed of stainless steel (Figure 3.9). Pressure gauges were rated up to 3000 psi as were the hoses that connected to the gas tank. The carbon dioxide gas tank was heated by an electrical heating element that automatically shut off at 60°C. Pressure vessels were place on an aluminum plate to evenly distribute the heat throughout the pressure vessels. Upon completion of the experiments, temperature and pressure were reduced and the slurry removed from the pressure vessels for further analysis (Figure 3.10).

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 3.9. Picture of the four pressure vessels used for all experiments. On top are the gauges which allow us to read the pressure inside the vessels. The vessels are sitting within an aluminum square plate on top of a hot plate. The purpose of the aluminum plate is to evenly distribute the heat from the hot plate to the entire vessel. Sticking out of the aluminum plate is a thermometer so that we can monitor the temperature of the vessels.



Figure 3.10. Image of the slurry from Experiment 7 after 3 months in the vessel.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

3.5 Methods

X-Ray Fluorescence (XRF)

Each of the three unreacted samples, (the DB, HBC and the HBM) were examined for both major and trace elemental analysis. Major elements analyzed (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, P) were obtained on fused La-bearing lithium borate glass disks using a Siemens MRS-400 multi-channel, simultaneous X-ray spectrometer. The analytical methods are modified from those of Norrish and Hutton (1969). Trace element data (Nb, Zr, Y, Sr, Rb, Th, Pb, Ga, Zn, Ni, Cr, V, Ba, Ce, La) are obtained using pressed powder pellets on a recently acquired Philips PW2400 sequential spectrometer. Intensities are corrected for non-linear background, interelement interferences and variations in mass absorption coefficients, using methods modified from those of Norrish and Chappel (1967). Mass absorption coefficients for elements with shorter characteristic wavelengths than the Fe-absorption edge are estimated from the intensity of the Compton radiation of the Rh X-ray tube (Reynolds, 1967). Mass absorption coefficients of elements with longer characteristic radiation than the Fe-absorption edge are calculated from the Compton-derived mass absorption coefficients, after allowance is made for Fe and Ti intensities (Walker, 1973).

X-Ray Diffraction (XRD)

Upon completion of each experiment, samples were removed from pressure vessels and placed in a 50°C drying oven for 24 hours to remove all water. Once dry, each sample was then reground using a mortar and pestle to achieve a uniform powder with which to run on the X-Ray Diffraction Analyzer. This is a non destructive technique which allows for the identification of crystallography structures based upon the diffraction of x-rays on the crystalline orientation of a given sample. All samples were run on the XRD prior to any experimentation in order to determine how much initial carbonate might be present. All samples were determined to have initial amounts of carbonate. This number was then subtracted from any carbonate numbers from experimented samples. Carbonate phases were determined by X-Ray Diffraction (XRD) with angle parameters of 13° - 67°, a step size of .020, and a time of 3 seconds per step. Abundance of carbonate minerals was determined by taking the intensity of each peak and dividing it by the reference intensity ratio (I/I_C), (Bayliss, 1986). The resulting number was then divided by the total reference intensity ratio of the entire sample and then multiplied by 100 to provide weight percent of carbonate produced. Carbonate minerals identified by XRD included calcite, dolomite, ankerite, siderite, aragonite, strontianite, witherite and vaterite.

Scanning Electron Microscope (SEM)

Scanning Electron Microscopy was used on selected experimented samples to attain a high resolution image of carbonate mineralization as well as pitting and dissolution associated with the chemical reaction (Figure 3.11). The SEM uses a high energy beam of electrons to scan a sample. These electrons interact with the atoms of the sample which in turn produce data concerning the samples topography and composition (Figure 3.12). With the SEM we are able to

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 3.11. A typical image taken with the Scanning Electron Microscope (SEM). This is an image of the HBM. Notice the green cross mark located roughly in the center of the image. The SEM is able to focus a beam onto that specific spot and give a chemical analysis of it.



Figure 3.12. The above spectrum is the chemical analysis of the cross mark figure 3.10.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

produce maps of specific elements, such as calcium, barium, or iron, on images of reacted samples. This type of imaging and mapping is helpful in that it identifies where carbonation is occurring. Pitting on mineral surfaces can also be observed on the images showing where the chemical reactions are occurring as well as the mineral involved in the reaction.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Chapter Four: Results

4.1 Rock Chemistry and Mineralogy

The Holyoke Basalt is made up of plagioclase phenocrysts in a fine grained groundmass of plagioclase, augite and pigionite (Philpotts et al., 1996). As seen in Figure 4.1, both Holyoke Basalt samples plot almost on top of one another because their composition is so similar. According to the Total Alkali and Silica (TAS) plot both Holyoke Basalts are basalts in the Tholeiitic field which means that they are sub alkaline and have lower amounts of sodium than other basalts (Figure 4.1). The TAS plot is divided into two sections; the upper left part of the plot is the alkaline domain while the low right section is the sub alkaline domain. The Deerfield Basalt is higher in sodium which is why it falls in the Hawaiite zone of the TAS diagram, just outside of the Tholeiitic domain.

Each sample has roughly 50% SiO₂ which is typical of basalts (Figure 4.2). Also, CaO, MgO, MnO and Fe_2O_3 concentrations are all quite similar throughout the three samples. These are the major cations that will react to form carbonate minerals. Interestingly Sr and Ba, the cations in strontianite and witherite, have different values across samples, with both Holyoke Basalt samples having higher amounts of each (Figure 4.2). Spider diagrams where the elemental chemistry of the basalts is compared to mid-ocean ridge basalt also indicate that the Holyoke basalt samples are very similar in composition whereas the Deerfield basalt is different, particularly with respect to K, Rb and Th content (Figure 4.3).

When we plot the different oxide concentrations which will react to form carbonate minerals against SiO_2 concentrations of each sample, an interesting pattern appears. CaO, MgO, Ba, and Sr concentrations are all higher in the Holyoke basalts (Figure 4.4). However, with MnO and Fe₂O₃ the difference between the three samples is much less apparent. Overall, this indicates the Holyoke basalts have higher concentrations of minerals bearing the cations which would react to form carbonate minerals.

4.2 Carbonate Production

The following is an experiment-by-experiment discussion concerning the production of carbonate. Results from XRD show production of carbonate in 22 of the 25 experiments (Figure 4.5) Total wt % change in carbonate ranges from -2% to 25% in the three hour experiments. In general, the HBN and HBC are more reactive than DB in most experiments. DB requires supercritical conditions (1400 psi and 100°C) and longer residence times to produce carbonate (experiments 1 and 7). Whereas HBM and HBC are capable of reacting with CO₂ at lower pressures (800 psi) and temperatures (20°C) (experiments 2,3,4). In addition to positive carbonate production, negative numbers are also reported. Unreacted samples were analyzed by XRD to quantity how much carbonate because they were precipitated after the basalts had formed. All three samples had varying amounts of secondary carbonates, with the Deerfield basalt having the highest amount. When the samples were then reacted some actually decreased in either the total carbonate or in specific carbonate minerals.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

TAS (Cox et al. 1979)



Figure 4.1. Total Alkali & Silica (TAS) graph. All three samples are plotted (small circles); however, the two Holyoke Basalt samples are so similar they appear as one data point. The Deerfield Basalt is plotted in the Hawaiite zone while both Holyoke Basalt samples are in the Tholeiitic Basalt zone.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Major & Trace Element Data for All Samples

Figure 4.2. Bar graph comparing the major and trace element concentrations of all three samples.



Figure 4.3. Spider diagram plotting the chemical composition of the Holyoke and Deerfield basalts against the chemical composition of mid ocean ridge basalts (MORB). The Y axis shows how enriched a sample is in a given element in relation to MORB chemical compositions. The Deerfield basalt is the line that is unlike the other two, more closely related, Holyoke basalts.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 4.4. Plots showing the major carbonate forming cations verses SiO_2 concentrations.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

51.60051.80052.00052.200

SiO2 (wt.%)

Geosciences Department University of Massachusetts 30

51.600 51.800 52.000 52.200

SiO2 (wt.%)



Figure 4.5. Bar graph representing all experiments conducted and the total change in carbonate produced. Refer to Figure 3.7 for the specific parameters of each experiment.

Experiment 1

Of all the experiments, experiment 1 was the least productive, producing less than 5% carbonate per sample. Of the three samples, the most productive sample was the Deerfield Basalt (DB) producing 4.4% ankerite and 0.7% strontianite (Figure 4.6). Following DB, the next most productive sample was HBM, again producing ankerite at 4.3% (Figure 4.7). HBG did not produce any ankerite in this experiment (Figure 4.8).

Experiment 2

Experiment 2 was conducted at low pressures and high temperatures, resulting in a gas phase. In this experiment DB produced almost no carbonate, only 0.2% in total (Figure 4.6). The Holyoke Basalt samples performed much better at 10% (HBM) and 15% (HBC) (Figures 4.7 and 4.8). In both Holyoke Basalt samples 5% of the total carbonate produced was calcite. This is the only experiment where two samples produced almost identical amounts of the same mineral. In HBM, the next most predominate mineral is dolomite at 5.3%. In HBC, ankerite is the next most predominate mineral at 6.5%. Throughout experiments 2 through 7 HBM will always precipitate appreciable amounts of dolomite while HBC will precipitate significant amounts of ankerite.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011


Figure 4.6. Bar graph showing different carbonate species produced in each experiment for the Deerfield Basalt. Negative numbers represent carbonate species lost due to geochemical reactions.



Figure 4.7. Bar graph showing carbonate species produced in each experiment for the Holyoke Basalt Massachusetts (HBM) sample. When the graph shows negative numbers this is showing carbonate that has been removed from the original sample through the experimentation process.



Figure 4.8. Bar graph showing carbonate species produced by Holyoke Basalt Connecticut (HBC) sample over all experiments. Negative numbers reflect carbonate minerals that were removed during experiments via geochemical reactions from the initial sample.

In experiment 3, temperatures were low and pressure was high, resulting in a liquid phase. Here, the DB did much better than in experiment 2, producing a total of 4% carbonate which is similar to that of experiment 1 (Figure 4.6). Again, the most produced mineral in DB is ankerite. The most productive sample in this experiment is the HBM at 16.6% with calcite and dolomite being the most dominant carbonate minerals (Figure 4.7). HBC produced very little carbonate in this experiment, a total of 1.6% (Figure 4.8). It appears that of the three samples, HBM is the only one that is capable of producing large amounts of carbonate in short time frames when CO_2 is at a liquid phase.

Experiment 4

Experiment 4 held both low pressures and temperatures resulting in a gas phase of CO₂. DB did very poorly in this experiment, reducing total carbonate by a -0.1% (Figure 4.6). However, both Holyoke Basalt samples did well with HBM producing a total 9% and HBC 15.9% (Figures 4.7 and 4.8). Again, each Holyoke Basalt sample produced their signatory carbonate mineral, ankerite in HBC and dolomite in HBM. However, this is the first experiment that witherite, a barium carbonate, is produced by HBC. Interestingly, witherite is produced in large quantities very quickly once it starts to precipitate.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

In experiment 5, CO₂ was again at a supercritical phase. The only difference here was the mass of sample and volume of water was increased by 50% from the original amounts in experiment 1. As previously stated, we were examining whether or not this increase would result in a linear increase in carbonate production. This was not at all what happened. In DB, total carbonate production actually decreased from experiment 1 by 0.6% (Figure 4.6). However, carbonate production in HBC and HBM increased many times over with HBM having the largest increase at 25.6% (Figures 4.7 and 4.8). Again, witherite seems to be associated with high carbonate production rates, specifically for HBM. HBC does produce some witherite at 5.7%, but HBM produced a total of 20.8%. It appears that once witherite begins to precipitate it happens very quickly and in large amounts. Why HBM produces more witherite than HBC may be a function of the barium concentration in the rock. HBC has 97 ppm of barium whereas HBM has 83 ppm (Figure 4.2).

Experiment 6

Experiment 6 was also at supercritical conditions, and similar to experiment 5, but differed in that the water to sample ratio was increased to 1/1. Again DB performed poorly with a negative carbonate number of 1.9% (Figure 4.6). Both Holyoke Basalt samples produced similar total carbonate percentages of 22.4% for HBM and 20.7% for HBC (Figures 4.7 and 4.8). This is the closest that they perform of any experiment. HBC produced 4.1% of ankerite and HBM produced 5.7% dolomite and both produced witherite, with HBC at 18.1% and HBM at 15.5%.

Experiment 7

In experiment 7 conditions were replicated that were identical to that of Schaef et al., (2009), except for the length of the experiment. Experiment 7 was conducted for 90 days, as opposed to the Schaef et al., (2009) work which lasted upwards of 800 days. In this experiment DB outperformed all of the samples producing a total of 8.1% carbonate (Figures 4.9 and 4.6). Interestingly, DB produced over 12% witherite, which is the largest amount of any carbonate mineral DB ever produced. HBM also produced a small amount of witherite at 2.6% (Figure 4.7). In their experiments, Schaef et al., (2009) found that the Holyoke basalt produced siderite (FeCO3) at 18 wt. % after 230 days and 72 wt % after 854 days and calcite (CaCO3) precipitation with ranges from 21.0 wt. % to 52.5 wt. %. Interestingly, in our work siderite was one of the least precipitated minerals in any of the experiments and was not produced at all in experiment 7. It is possible that because our experiments were of a much shorter duration than Schaef et al., (2009) siderite was not provided the necessary time to precipitate.

4.3 Summary of Results

Of all three samples, HBC and HBM consistently produced more carbonate, except in the longest duration experiment, experiment 7. During each experiment, each sample produced a unique combination of carbonate minerals. However, it seems that once witherite appeared in a sample,

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 4.9. SEM image of DB sample after 3 months. Calcium carbonate minerals have formed on the edge of this plagioclase crystal. The carbonate crystals are the small white angular pieces along the edge of the larger grey plagioclase crystal.

total carbonate production increased rapidly. In addition, the appearance of witherite and its concentrations cannot be predicted by the total amount of barium in a given sample. Different carbonate species are being precipitated under different conditions. This does not seem to be completely dependent upon the specific mineralogy of a given sample. For example, Figure 4.10 is a comparison between HBC and HBM of the major cations which make up ankerite and dolomite, the signatory carbonate minerals of each. Notice the very minor differences between each and yet HBM consistently produces dolomite while HBC produces ankerite.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| | HBC (wt %) | HBM (wt. %) | Difference |
|--------------------------------|------------|-------------|------------|
| CaO | 10.229 | 10.207 | .022 |
| MgO | 5.729 | 5.931 | .202 |
| MnO | .244 | .243 | .001 |
| Fe ₂ O ₃ | 13.802 | 13.955 | .153 |

Figure 4.10. Table showing the concentrations of different oxides in each of the three samples.

Another important question that we attempted to answer was from what minerals are the cations (Ca, Mn, Mg, Fe) coming from? As stated earlier, these basalts are predominantly made up of pyroxenes and plagioclases. Pyroxene had the general formula $XY(Si, Al)_2O_6$ where X represents calcium, sodium, iron²⁺, and manganese and Y represents smaller ions such as chromium, iron ³⁺ and aluminum. Plagioclase has varying amounts of sodium or calcium with the following general formula, (Na, Ca) $AlSi_3O_8$. Therefore, it is possible that the calcium could come from either mineral whereas with the Mg, Mn, and Fe it is more likely that it came from the pyroxene.

To better attain an answer to this question an experiment was designed where SEM images were taken of a highly polished sample before it was placed in the pressure vessel. After submitting the polished sample to testing, the surface of the sample was imaged again on the SEM (Figure 4.11). The conditions for this SEM experiment were identical to that of experiment 4 (800 psi and 20°C). The difference, however, was that a smooth slice of rock was used rather than a powder. Images after the experiment show intensive degradation of the sample surface (Figure 4.12). However, the pitting was so severe that it was impossible to determine what the original composition of the mineral was. This severity in pitting indicates that the dissolution of CO_2 into the water occurs quickly resulting in the formation of carbonic acid. This acidification is what is causing the pitting and thus the release of cations necessary for carbonation. Clearly, alteration is occurring in as little as three hours.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Figure 4.11. SEM image of DB slide before reaction.



Figure 4.12. SEM image of *DB* slide after 3 hour reaction. The surface has a covering of some type of clay mineral that was not there prior to experimentation. Notice in the bottom left corner the pitting of the crystals.

Chapter Five: Summary and Conclusions

The results of this study suggest that the Holyoke and Deerfield basalts of the Connecticut River Valley have potential as storage units for carbon dioxide, specifically when an ex-situ mineralization scheme is considered. From this work, it can be concluded that supercritical carbon dioxide is not necessary to promote carbonate mineralization. Due to the high cost associated with compressing CO₂ from a gas to a supercritical fluid, this finding has important implications in future economic analysis of CCS projects. Another major conclusion of this work is that carbonate precipitation can occur rapidly, in as short as three hours, with small grain sizes. Furthermore, each sample precipitates its own unique suite of carbonate minerals. This is true even for samples that are within the same geologic unit. Given this finding it is important for any future basalt carbon sequestration investigations to consider how minor shifts in geochemical composition, as well as geographic location, can impact mineralization products and rates. The formation of Witherite (BaCO₃) has proven to be an important carbonate mineral in the sequestration of CO₂. Once witherite is formed it appears to precipitate very quickly. In almost all experiments that exceeded a 15% increase in total carbonate, witherite is the most abundant carbonate mineral produced.

This work shows that when designing the optimal conditions for carbonate precipitation it is important to look at each location individually. For the Holyoke basalt samples, an increase in water increased carbonate production. However, this increase in water had the opposite effect on the Deerfield basalt where initial carbonate in the sample was actually dissolved due to the increase in carbonic acid. However, it would be interesting to test whether the increase in water would eventually initiate carbonate precipitation had the experiment on DB been allowed to run for a longer period of time. Both Holyoke samples also performed well when CO₂ was in gas form, either at low pressure and high temperatures or at both low pressures and temperatures. Future investigations would benefit from an experiment where both temperatures and pressures were below supercritical and the water/sample ratio was increased to 1/1. This experiment would combine elements of the two highest performing experiments of this work. Optimal conditions for the Deerfield basalt involved being exposed to supercritical conditions for extended periods of time, at least 3 months. The optimal conditions for carbonate mineralization as a sequestration strategy will always need to be determined through initial experimental work on a site by site basis. Future work needs to examine why similar basalts produce different suites of carbonate minerals. This information will further our understanding and ability to predict amounts of carbonate precipitated.

Finally, when using this work to examine basalts from around the world as candidates for carbonate mineralization desirable characteristics need to include;

Fine grain matrix
> 80 ppm Barium (Ba) content
Tholeiitic Basalt with ~50% SiO₂

In order to create a more thorough list of necessary characteristics more research is needed that would first test for these characteristics in basalts at other locations.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Chapter Six: Implication of Results and Next Steps

Half of all electricity in the United States is generated by coal combustion, which, in turn, produces over 30% of CO₂ emissions. In order to reduce the rate of climate change it is necessary to address these emissions. One method of reducing CO₂ emission is through carbon capture and sequestration (CCS). Most CCS research and pilot projects have focused on the capture, transport and geologic sequestration of CO₂ in the subsurface. A few researchers, however, have explored the idea of an ex-situ mineralization process which converts CO₂ into a solid carbonate mineral in a reactor above ground (Lackner et al., 1997; O'Conner et al., 2002; Gerdemann et al., 2007; Oelkers et al., 2008). In each of these proposed ex-situ processes a pure mineral substrate and either water, or a bicarbonate/salt slurry, was used to sequester the CO₂ (Lackner et al., 1997; O'Conner et al., 2002; Gerdemann et al., 2007.,). Our work, however, has shown favorable data from experiments that explore basalts as the feed stock for an ex-situ mineralization process, rather than a solitary mineral. In addition, these new experiments show that conversion of CO₂ to stable carbonate occurs quickly (within 3 hours) and under non-supercritical conditions, which has implications for either an ex-situ or in-situ application.

The geologic storage of captured carbon dioxide has many benefits when considering the need for immediate deployment of this technology. From the oil and gas industry, much is understood about reservoir characterization, well technology and pipelines. CCS, however, unlike oil and gas extraction involves the placement of a substance underground for an infinite amount of time. Thus, CCS is challenged by modeling proposed reservoirs for possible leakage pathways as well as developing monitoring techniques that can withstand the test of time. An ex-situ carbonate mineralization process has the advantage that once the CO_2 is locked in the minerals, there is no need for extensive monitoring or modeling. Because the CO_2 is locked in minerals, it is possible that this type of a sequestration process will gain greater amounts of public acceptance in a shorter amount of time. In addition, an ex-situ mineralization process does not require the transport of CO_2 in an extensive pipeline network. The basalt could be delivered to the power plant and then removed for reuse once reacted.

Challenges to an ex-situ carbon mineralization process include the large scale of such an effort, the need to increase efficiency through increasing the rate of carbonate mineralization, and cost (Oelkers et al., 2008). In regards to the necessary scale of an effective ex-situ project, it is important to highlight the enormity of any carbon sequestration project, above or below the ground, due to the large amount of emissions produced. The relative cost of an ex-situ reactor has been explored (Huijgen et al., 2007) and determined that the major costs of an ex-situ reactor are associated with the feed stock and electricity for grinding. However, this exploration has used a single mineral, such as wollastonite, rather than exploring the possibility of using a mixed mineral host, like basalt. Additionally, Huijgen et al., (2007) only examined the cost of the process post compression, or once the CO_2 was in a supercritical phase.

It is generally understood that the major costs associated with CCS are within the capturing of CO_2 , which includes the compression of CO_2 from a gas to a supercritical phase. One of the major findings of this work is that it is not necessary to have CO_2 in a supercritical phase to produce carbonate minerals. Therefore, this finding combined with the conclusion that it is

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

possible to use basalt, a mixed mineral host, for carbonate mineralization have important implications in future energy and life cycle analysis for an ex-situ reactor. Furthermore, once the basalt is reacted, there is the potential for it to be reused in various applications, such as in road construction.

6.1 Next Steps

The favorable results from this work necessitate further investigation into a variety of topics related to mineral sequestration. The following list describes suggested actions to be taken in order to attain whether an ex-situ mineral reactor process is feasible. It is also important to determine whether an in-situ geologic storage program is possible in the basalts of the Connecticut River Valley.

1. Energy and Life Cycle Analysis

The process of carbon capture and storage (CCS) involves energy at every stage of the project. Leading CCS networks (Global CCS Institute, IEA-GHG, EU ZEP) have compiled resources which examine the overall costs associated with each step of a traditional CCS project. Huijgen et al., (2007) has produced an economic analysis of an ex-situ mineralization reactor which utilizes a solitary mineral as the reaction host citing the cost of attain the mineral as extremely high. Thus, to date, no energy and life cycle analysis has been conducted on an ex-situ mineralization process that utilizes basalt as the host for geochemical reactions. In addition, as this work has shown, it is necessary to also consider the financial implications of an ex-situ mineralization scheme where supercritical carbon dioxide is not necessary.

2. Small Scale Pilot Study

Our work has shown favorable data for sequestration under a variety of conditions. Next, these conditions need to be explored on a larger scale using larger amounts of material and reactors. Additionally, it will be important to understand how these reactions change when CO_2 is flowing through a reactor rather than just being sedentary once pumped into it. When CO_2 exits a power generation facility through a smoke stack it does so with a variety of other gases, usually referred to as SOX (sulfur oxides) and NOX (nitrous oxides). It is this combination of gases that warrants the CO_2 capture process to purify the stream to just CO_2 . To further reduce costs associated with capture, it would be useful to conduct research where the CO_2 stream is not a pure one where sulfur and nitrous oxides were present to better understand if a reduced purification process would still result in carbonation.

3. Connecticut River Valley Basalt Study

To date, there is a lack of information on the geology of the Connecticut River Valley at depth. What is known has been determined using various geological techniques that allow us to infer what is occurring at depth. Regardless of whether an ex-situ or an in-situ carbon sequestration program is considered, it is important to know how much basalt there is and where exactly it is. An important finding of this work is that even within the same geologic unit, as in the Holyoke

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

basalt, geochemical reactions vary. Therefore, more samples need to be collected and examined for sequestration potential at depth for a given unit.

4. Global Basalt Study

As stated earlier, basalts exist on every continent on earth, as well as in the oceans. There are many regions where suitable deep saline aquifers do not exist for CCS. Therefore, if CCS is to be considered as a major carbon dioxide reducing strategy it is important that each region have the ability to utilize CCS. By further exploring the potential for basalts to sequester CO_2 the regions that can implement CCS increases. When an ex-situ strategy is considered, CCS technology is no longer determined by local or regional geology but could be applied anywhere. In the interest of being able to apply CCS to the global community, it is necessary to examine the basalts of the world to find which rocks respond similarly to the Holyoke basalt when reacted with CO_2 and water.

6.2 Carbonate Mineralization and the State of Massachusetts

The State of Massachusetts has put itself forward as a leader of the nation in climate change policy and green work force development. This is evident through policies such as the creation of the Global Warming Solutions Act, the Green Communities Act, and the formation of the Massachusetts Clean Energy Center. Carbonate mineralization in basalts, as a form of carbon capture and sequestration, is another opportunity for Massachusetts to carry on this legacy. Currently, the State is pursuing the development of many renewable energy sources to help reduce the total amount of greenhouse gases produced by the state. Massachusetts has opportunities for wind power, both in the eastern and western parts of the state, solar power, as well as geothermal potential. However, it will be difficult for the State to produce all of the energy it needs through renewable resources. As such, CCS projects can help reduce the amount of carbon dioxide emissions in Massachusetts while still generating the power it needs. CCS projects are often criticized as requiring too much capital investment to make them viable. These assessments, however, are only true when a CCS project is compared to solar, wind, or even nuclear with the current subsidies attached.

As shown in Figure 6.1, when subsidies are removed from low carbon energy technologies CCS becomes competitive. Power generation facilities with CCS are actually cheaper than solar and offshore wind, both of which Massachusetts is developing actively.

The potential for Massachusetts to sequester carbon dioxide lies in basalts. These are basalts are part of a larger chain of basalts known as the Newark Supergroup that stretches from Newfoundland to Georgia. Massachusetts' climate policies and initiatives combined with its outstanding academic and research facilities offer a unique opportunity to take advantage of this geologic resource.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



 $_{\rm E\,/\,MWh}$ $\,$ CoE - Low Carbon Technologies Ordered in 2011-2016 Period $\,$

Source : Alstom analysis 2011. CCS 2015 costs, including on shore T&S and CO_2 price (Flue Gas Recirculation case for CCS Gas CC, average Amine/Oxy for CCS Hardcoal)

Figure 6.1. Graph comparing the Cost of Energy (CoE) of low carbon technologies with NO subsidies attached. The two technologies highlighted in green are power facilities with CCS. The one to the left is a combined cycle gasification technology (CCGT) CCS plant, which is essentially a natural gas power plant. The one to the right is a coal power plant with CCS.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

References Cited

- 2008 Carbon Sequestration Atlas of the United States and Canada, 2nd edition (2008) United States Department of Energy, Office of Fossil Energy, National Energy Technology Laboratory. 142 pgs.<u>http://www.netl.doe.gov/technologies/carbon_seq/refshelf/atlasII/</u>
- Alstom (2011) Cost assessment of fossil power plants equipped with CCS under typical scenarios. POWER-GEN Europe, Milan, Italy 7-9 July 2011.
- Alfredsson, H. A., Hardarson, B. S, Franzson, H. Gislason, S. R., (2008) CO2 sequestration in basaltic rock at the Hellisheidi site in SW Iceland: stratigraphy and chemical composition of the rocks at the injection site. *Mineralogical Magazine* vol.71, 1-4.
- Bayliss, P., (1986) Quantitative Analysis of Sedimentary Minerals by Powder X-Ray Diffraction. *Powder Diffraction* vol. 1, 37pp.
- Charan, S. N., Begum, Zahida, (2009) Geological sequestration of carbon dioxide in Deccan basalts: preliminary laboratory study. *Current Science* vol. 96, 288-291.
- Cox, K. G.; Bell, J. D.; Pankhurst, R. J. (1979) The interpretation of Igneous Rocks. *George Allen & Unwin : London, United Kingdom.*
- *The Costs of CO*₂ *Capture, Transport and Storage*.(2011) EU Zero Emissions Platform. <u>http://www.zeroemissionsplatform.eu/library/publication/165-zep-cost-report-</u> <u>summary.html</u>
- Flaathen, T.K., Gislason, S.R., Oelkers, Eric H., Sveinbjornsdottir, Arny E., (2009)Chemical evolution of the Mt. Hekla, Iceland, groundwaters: A natural analogue for CO(2) sequestration in basaltic rocks. *Applied Geochemistry* vol.24, 463-474.
- Froelich, A.J., Robinson, Jr., G.R. (1988) Studies of the Early Mesozoic Basins of the Eastern United States. U.S. Geological Survey Bulletin 1776.423 pp.
- Gerdemann, S.J., O'Connor, W., Dahlin, D., Penner, L., Rush, H., (2007) Ex Situ Aqueous Mineral Carbonation. *Environmental Science Technology* **41** 2587-2593.
- Goldberg, D.S., Takahashi, T., Slagle, A.L. (2008) Carbon dioxide sequestration in deep-sea basalt. *Proceedings of the National Academy of Science***105**, 9920-9925.
- Gysi, A. P. Stefansson, A., (2008) CO(2)-basalt interaction Numerical simulation and experimental study. *Geochimica et Cosmochimica Acta*. vol.71, A894-A894.
- Huijgen, Wouter J. J., Comans, Rob N. J. Witkamp, Geert-Jan. (2007) Cost evaluation of CO2 sequestration by aqueous mineral carbonation. *Energy Conversion and Management*. vol.48 1923-1935.
- IPCC, 2005: IPCC Special Report on Carbon Dioxide Capture and Storage. Prepared by Working Group III of the Intergovernmental Panel on Climate Change [Metz, B., O. Davidson, H. C. de Coninck, M. Loos, and L. A. Meyer (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 442 pp. <u>http://www1.ipcc.ch/pdf/special-reports/srccs/srccs_wholereport.pdf</u>

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

- Lackner, Klaus S., et al. "Carbon Dioxide Disposal in Carbonate Minerals." *Energy* 20.11 (1995): 1153-70.
- Matter, Juerg M., et al. "Permanent Carbon Dioxide Storage into Basalt: The CarbFix Pilot Project, Iceland." *Energy Procedia* 1.1 (2009): 3641-6.
- Matter, J., Kelemen, P., (2009) Permanent storage of carbon dioxide in geological reservoirs by mineral carbonation. *Nature Geoscience* **2**, doi: 10.1038/NGEO683
- McGrail, B.P., Schaef, H.T., Ho, A.M., Chien, Y.J., Dooley, J.J., Davidson, C.L. (2006) Potential for carbon dioxide sequestration in flood basalts. *Journal of Geophysical Research – Solid Earth* **111**, B12201.doi:10.1029/2005JB004169.
- Norrish, K. and B. W. Chappell. (1967). X-Ray Fluorescence Spectrography. In *Physical Methods in Determinative Mineralogy*, edited by J. Zussman, pp. 161-214. Academic Press, New York, New York.
- Norrish K. and Hutton JT., (1969) An Accurate X-Ray Spectrographic Method for Analysis of a Wide Range of Geological Samples. *Geochimica et Cosmochimica Acta*. vol.33 doi 10.1016/0016-7037(69)90126-4
- O'Connor, W. K., Dahlin, D. C., Rush, G. E., Gerdemann, S. J. & Nilsen, D. N. Aqueous Mineral Carbonation: Final Report DOE/ARC-TR-04-002 (US Department of Energy,) 2004.
- Oelkers,, E.H., Gislason, S.R., Matter, J. (2008) Mineral carbonation of CO₂. *Elements* **4**, 333-337.
- Philpotts, AR., Carroll, M., Hill, JM., (1996) Crystal-mush compaction and the origin of pegmatitic segregation sheets in a thick flood-basalt flow in the Mesozoic Hartford basin, Connecticut. *Journal of Petrology* vol. 37, 811-836.
- Philpotts, A.R. (1998) Nature of a flood-basalt-magma reservoir based on the compositional variation in a single flood-basalt flow and its feeder dike in the Mesozoic Hartford basin, Connecticut. *Contribution to Mineralogy and Petrology***133**, 69-82.
- Reynolds RC., (1967) Estimation of mass absorption coefficient by Compton scattering: Improvements and extensions of the method. Amer. Mineral. 52, 1493-15m.
- Schaef, H. T, McGrail, B. P., Owen, A. T., (2008) Carbonate mineralization of volcanic province basalts. *International Journal of Greenhouse Gas Control* doi 10.1016/j.ijggc.2009.10.009.
- Schaef, H. T., B. P. McGrail, and A. T. Owen. "Basalt- CO2–H2O Interactions and Variability in Carbonate Mineralization Rates." *Energy Procedia* 1.1 (2009): 4899-906.
- Walker, D., (1973) Behavior of X-ray Mass Absorption Coefficients near Absorption Edges: Reynolds' Method Revisited. *American Mineralogist*, Volume 58, pages 1069-1072,

Walsh, M.P. (2008) Petrology and Provenance of the Triassic Sugarloaf Arkose, Deerfield Basin, Massachusetts: M.S. thesis, Department of Geosciences, University of Massachusetts Amherst, Amherst, MA, 270 p.

Appendix A: X-Ray Diffraction (XRD) Data

The following tables are all of the data from XRD analysis divided by experiment. The first column is the mineral identified based on its reference peak number (the second column). Columns 4 and 5 are based off of Bayliss (1989) to determine the total abundance of the given mineral as a percent. Column 6 gives the total abundance of that mineral. The following column, column 7 gives the abundance of that mineral in the unreacted samples. Finally, the last column offers the difference in mineral abundance between the unreacted sample and the reacted sample. Data from the unreacted samples follows experiment 7 in this Appendix.

Experiment 1

| DB 3hour | | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|--|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change | |
| Plagioclase | 3.19 | 100 | 58.82 | 70.7 | 69.1 | 1.6 | |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 | |
| Pyroxene | 3 | 11.21 | 11.21 | 13.5 | 17.0 | -3.5 | |
| Quartz | 3.34 | 1.98 | 0.46 | 0.6 | 3.4 | -2.8 | |
| Clay | 4.5 | 1.63 | 0.63 | 0.8 | 0.7 | 0.1 | |
| Calcite | 3.03 | 15.8 | 4.27 | 5.1 | 5.4 | -0.3 | |
| Ankerite | 2.9 | 10.16 | 3.63 | 4.4 | 0.0 | 4.4 | |
| Dolomite | 2.89 | 3.74 | 1.34 | 1.6 | 1.6 | 0.0 | |
| Siderite | 2.8 | 0.96 | 0.34 | 0.4 | 0.5 | -0.1 | |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 | |
| Strontianite | 3.54 | 9.36 | 2.53 | 3.0 | 2.3 | 0.7 | |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0.0 | 0.0 | |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.3 | -0.3 | |
| | | SUM | 83.23 | 100.0 | | | |

| HBG 3HOUR | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 87.1 | 71.9 | 15.2 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 1.59 | 1.59 | 2.4 | 16.4 | -14.0 |
| Quartz | 3.34 | 0 | 0.00 | 0.0 | 2.6 | -2.6 |
| Clay | 4.5 | 3.11 | 1.20 | 1.8 | 2.2 | -0.4 |
| Calcite | 3.03 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Ankerite | 2.9 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Dolomite | 2.89 | 13.05 | 4.66 | 6.9 | 6.9 | 0.0 |
| Siderite | 2.8 | 1.64 | 0.59 | 0.9 | 0 | 0.9 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 2.39 | 0.65 | 1.0 | 0 | 1.0 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 67.51 | 100.0 | | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| HBN 3HOUR | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 84.2 | 64.1 | 20.1 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 4.48 | 4.48 | 6.4 | 28.2 | -21.8 |
| Quartz | 3.34 | 0 | 0.00 | 0.0 | 1.4 | -1.4 |
| Clay | 4.5 | 1.45 | 0.56 | 0.8 | 0.6 | 0.2 |
| Calcite | 3.03 | 1.13 | 0.31 | 0.4 | 0 | 0.4 |
| Ankerite | 2.9 | 16.03 | 5.73 | 8.2 | 3.9 | 4.3 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 1.8 | -1.8 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 69.89 | 100.0 | | |

| DB pressure | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 74.4 | 69.1 | 5.3 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 11.24 | 11.24 | 14.2 | 17.0 | -2.8 |
| Quartz | 3.34 | 4.03 | 0.94 | 1.2 | 3.4 | -2.2 |
| Clay | 4.5 | 1.45 | 0.56 | 0.7 | 0.7 | 0.0 |
| Calcite | 3.03 | 10.3 | 2.78 | 3.5 | 5.4 | -1.9 |
| Ankerite | 2.9 | 2.87 | 1.03 | 1.3 | 0.0 | 1.3 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 | 1.6 | -1.6 |
| Siderite | 2.8 | 0 | 0.00 | 0.5 | 0.5 | 0.0 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 10.23 | 2.76 | 3.5 | 2.3 | 1.2 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Vaterite | 3.29 | 3.55 | 0.96 | 1.2 | 0.3 | 0.9 |
| | | SUM | 79.09 | 100.5 | | |

| HBC pressure | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 44.4 | 71.9 | -27.5 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 34.64 | 34.64 | 26.1 | 16.4 | 9.7 |
| Quartz | 3.34 | 40.67 | 9.46 | 7.1 | 2.6 | 4.5 |
| Clay | 4.5 | 3.92 | 1.51 | 1.1 | 2.2 | -1.1 |
| Calcite | 3.03 | 26.32 | 7.11 | 5.4 | 0 | 5.4 |
| Ankerite | 2.9 | 24.21 | 8.65 | 6.5 | 0 | 6.5 |
| Dolomite | 2.89 | 11.67 | 4.17 | 3.1 | 6.9 | -3.8 |
| Siderite | 2.8 | 0 | 0.00 | 0.5 | 0 | 0.5 |
| Aragonite | 3.4 | 5.55 | 5.55 | 4.2 | 0 | 4.2 |
| Strontianite | 3.54 | 5.93 | 1.60 | 1.2 | 0 | 1.2 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 3.55 | 0.96 | 0.7 | 0 | 0.7 |
| | | SUM | 132.48 | 100.5 | | |

HBM pressure Mineral Ref. Peak (Å) Intensity Intensity / WF Abundance (%) Abundance in Unreacted Total Change Plagioclase 3.19 100 58.82 41.6 64.1 -22.5 Amphibole 0.00 0.0 0 0.0 8.5 0 Pyroxene 3 53.55 53.55 37.8 28.2 9.6 Quartz 3.34 17.03 3.96 2.8 1.4 1.4 0.6 0.2 Clay 4.5 3.01 1.16 0.8 Calcite 3.03 26.32 7.11 5.0 0 5.0 3.9 Ankerite 2.9 22.59 8.07 5.7 1.8 Dolomite 2.89 21.13 7.55 5.3 0 5.3 3.55 Siderite 2.8 1.27 0.5 1.8 -1.3 Aragonite 3.4 0 0.00 0.0 0 0.0 0 Strontianite 3.54 0.00 0.0 0 0.0 Witherite 3.72 0 0.00 0.0 0 0.0 Vaterite 3.29 0 0.00 0.0 0 0.0 SUM 141.49 99.6

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| DB temperature | | | | | | |
|----------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 68.8 | 69.1 | -0.3 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 12.67 | 12.67 | 14.8 | 17.0 | -2.2 |
| Quartz | 3.34 | 6.19 | 1.44 | 1.7 | 3.4 | -1.7 |
| Clay | 4.5 | 1.95 | 0.75 | 0.9 | 0.7 | 0.2 |
| Calcite | 3.03 | 10.2 | 2.76 | 3.2 | 5.4 | -2.2 |
| Ankerite | 2.9 | 13 | 4.64 | 5.4 | 0.0 | 5.4 |
| Dolomite | 2.89 | 5.01 | 1.79 | 2.1 | 1.6 | 0.5 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 0.5 | -0.5 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 9.69 | 2.62 | 3.1 | 2.3 | 0.8 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.3 | -0.3 |
| | | SUM | 85.49 | 100.0 | | |

| HBC temperature | | | | | | |
|-----------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 52.8 | 71.9 | -19.1 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 34.84 | 34.84 | 31.3 | 16.4 | 14.9 |
| Quartz | 3.34 | 29.26 | 6.80 | 6.1 | 2.6 | 3.5 |
| Clay | 4.5 | 3.22 | 1.24 | 1.1 | 2.2 | -1.1 |
| Calcite | 3.03 | 6.06 | 1.64 | 1.5 | 0 | 1.5 |
| Ankerite | 2.9 | 17.27 | 6.17 | 5.5 | 0 | 5.5 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 | 6.9 | -6.9 |
| Siderite | 2.8 | 1.69 | 0.60 | 0.5 | 0 | 0.5 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 4.45 | 1.20 | 1.1 | 0 | 1.1 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 111.32 | 100.0 | | |

| HBM temperature | | | | | | |
|-----------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 54.2 | 64.1 | -9.9 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 16.52 | 16.52 | 15.2 | 28.2 | -13.0 |
| Quartz | 3.34 | 32.04 | 7.45 | 6.9 | 1.4 | 5.5 |
| Clay | 4.5 | 3.95 | 1.53 | 1.4 | 0.6 | 0.8 |
| Calcite | 3.03 | 32.73 | 8.85 | 8.2 | 0 | 8.2 |
| Ankerite | 2.9 | 18.18 | 6.49 | 6.0 | 3.9 | 2.1 |
| Dolomite | 2.89 | 21.42 | 7.65 | 7.0 | 0 | 7.0 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 1.8 | -1.8 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 4.45 | 1.20 | 1.1 | 0 | 1.1 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 108.51 | 100.0 | | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| DB 3hour NON SC | | | | | | |
|-----------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 74.5 | 69.1 | 5.4 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 10.73 | 10.73 | 13.6 | 17.0 | -3.4 |
| Quartz | 3.34 | 4.98 | 1.16 | 1.5 | 3.4 | -1.9 |
| Clay | 4.5 | 1.49 | 0.58 | 0.7 | 0.7 | 0.0 |
| Calcite | 3.03 | 11.65 | 3.15 | 4.0 | 5.4 | -1.4 |
| Ankerite | 2.9 | 2.61 | 0.93 | 1.2 | 0.0 | 1.2 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 | 1.6 | -1.6 |
| Siderite | 2.8 | 3.05 | 1.09 | 1.4 | 0.5 | 0.9 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 9.11 | 2.46 | 3.1 | 2.3 | 0.8 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.3 | -0.3 |
| | | SUM | 78.92 | 100.0 | | |

| HBC 3hour NON SC | | | | | | |
|------------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 38.7 | 71.9 | -33.2 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 49.63 | 49.63 | 32.7 | 16.4 | 16.3 |
| Quartz | 3.34 | 31.25 | 7.27 | 4.8 | 2.6 | 2.2 |
| Clay | 4.5 | 4.14 | 1.60 | 1.1 | 2.2 | -1.1 |
| Calcite | 3.03 | 6.34 | 1.71 | 1.1 | 0 | 1.1 |
| Ankerite | 2.9 | 19.03 | 6.80 | 4.5 | 0 | 4.5 |
| Dolomite | 2.89 | 22.33 | 7.98 | 5.2 | 6.9 | -1.7 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Witherite | 3.72 | 18.2 | 18.20 | 12.0 | 0 | 12.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 152.00 | 100.0 | | |

| HBM 3hour NON SC | | | | | | |
|------------------|---------------|-----------|----------------|---------------|------------------------|-----------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Carbonate |
| Plagioclase | 3.19 | 100 | 58.82 | 58.5 | 64.1 | -5.6 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 21.04 | 21.04 | 20.9 | 28.2 | -7.3 |
| Quartz | 3.34 | 25.57 | 5.95 | 5.9 | 1.4 | 4.5 |
| Clay | 4.5 | 0 | 0.00 | 0.0 | 0.6 | -0.6 |
| Calcite | 3.03 | 7.9 | 2.14 | 2.1 | 0 | 2.1 |
| Ankerite | 2.9 | 14.32 | 5.11 | 5.1 | 3.9 | 1.2 |
| Dolomite | 2.89 | 21.04 | 7.51 | 7.5 | 0 | 7.5 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 1.8 | -1.8 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 100.57 | 100.0 | | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| DB mass | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 71.5 | 69.1 | 2.4 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 10.25 | 10.25 | 12.5 | 17.0 | -4.5 |
| Quartz | 3.34 | 5.36 | 1.25 | 1.5 | 3.4 | -1.9 |
| Clay | 4.5 | 1.32 | 0.51 | 0.6 | 0.7 | -0.1 |
| Calcite | 3.03 | 19.57 | 5.29 | 6.4 | 5.4 | 1.0 |
| Ankerite | 2.9 | 9.03 | 3.23 | 3.9 | 0.0 | 3.9 |
| Dolomite | 2.89 | 2.46 | 0.88 | 1.1 | 1.6 | -0.5 |
| Siderite | 2.8 | 0.96 | 0.34 | 0.4 | 0.5 | -0.1 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 6.29 | 1.70 | 2.1 | 2.3 | -0.2 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.3 | -0.3 |
| | | SUM | 82.27 | 100.0 | | |

| HBC mass | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| | | | | | | |
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 51.9 | 71.9 | -20.0 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 31.36 | 31.36 | 27.7 | 16.4 | 11.3 |
| Quartz | 3.34 | 7.09 | 1.65 | 1.5 | 2.6 | -1.1 |
| Clay | 4.5 | 3.7 | 1.43 | 1.3 | 2.2 | -0.9 |
| Calcite | 3.03 | 10.32 | 2.79 | 2.5 | 0 | 2.5 |
| Ankerite | 2.9 | 7.32 | 2.61 | 2.3 | 0 | 2.3 |
| Dolomite | 2.89 | 2.46 | 0.88 | 0.8 | 6.9 | -6.1 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Aragonite | 3.4 | 5.7 | 5.70 | 5.0 | 0 | 5.0 |
| Strontianite | 3.54 | 5.62 | 1.52 | 1.3 | 0 | 1.3 |
| Witherite | 3.72 | 6.47 | 6.47 | 5.7 | 0 | 5.7 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 113.23 | 100.0 | | |

| HBM mass | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 38.3 | 64.1 | -25.8 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 35.37 | 35.37 | 23.0 | 28.2 | -5.2 |
| Quartz | 3.34 | 21.26 | 4.94 | 3.2 | 1.4 | 1.8 |
| Clay | 4.5 | 5.16 | 1.99 | 1.3 | 0.6 | 0.7 |
| Calcite | 3.03 | 5.68 | 1.54 | 1.0 | 0 | 1.0 |
| Ankerite | 2.9 | 24.42 | 8.72 | 5.7 | 3.9 | 1.8 |
| Dolomite | 2.89 | 24.63 | 8.80 | 5.7 | 0 | 5.7 |
| Siderite | 2.8 | 4.42 | 1.58 | 1.0 | 1.8 | -0.8 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Witherite | 3.72 | 32 | 32.00 | 20.8 | 0 | 20.8 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 153.76 | 100.0 | | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| DB Water | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 73.9 | 69.1 | 4.8 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 13.46 | 13.46 | 16.9 | 17.0 | -0.1 |
| Quartz | 3.34 | 4.74 | 1.10 | 1.4 | 3.4 | -2.0 |
| Clay | 4.5 | 0 | 0.00 | 0.0 | 0.7 | -0.7 |
| Calcite | 3.03 | 11.94 | 3.23 | 4.1 | 5.4 | -1.3 |
| Ankerite | 2.9 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Dolomite | 2.89 | 5.08 | 1.81 | 2.3 | 1.6 | 0.7 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 0.5 | -0.5 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 4.49 | 1.21 | 1.5 | 2.3 | -0.8 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.3 | -0.3 |
| | | SUM | 79.64 | 100.0 | | |

| HBC Water | | | | | | |
|--------------|---------------|-----------|----------------|---------------|-----------------------|----------------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundace in Unreacted | Total Change in Comp |
| Plagioclase | 3.19 | 100 | 58.82 | 47.1 | 71.9 | -24.8 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 27.53 | 27.53 | 22.0 | 16.4 | 5.7 |
| Quartz | 3.34 | 12.45 | 2.90 | 2.3 | 2.6 | -0.3 |
| Clay | 4.5 | 3.23 | 1.25 | 1.0 | 2.2 | -1.2 |
| Calcite | 3.03 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Ankerite | 2.9 | 14.33 | 5.12 | 4.1 | 0.0 | 4.1 |
| Dolomite | 2.89 | 12.23 | 4.37 | 3.5 | 6.9 | -3.4 |
| Siderite | 2.8 | 3.15 | 1.13 | 0.9 | 0.0 | 0.9 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 4.5 | 1.22 | 1.0 | 0.0 | 1.0 |
| Witherite | 3.72 | 22.58 | 22.58 | 18.1 | 0.0 | 18.1 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| | | SUM | 124.90 | 100.0 | | |

| HBC Water | | | | | | |
|--------------|---------------|-----------|----------------|---------------|-----------------------|----------------------|
| | | | | | | |
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundace in Unreacted | Total Change in Comp |
| Plagioclase | 3.19 | 100 | 58.82 | 47.1 | 71.9 | -24.8 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 27.53 | 27.53 | 22.0 | 16.4 | 5.7 |
| Quartz | 3.34 | 12.45 | 2.90 | 2.3 | 2.6 | -0.3 |
| Clay | 4.5 | 3.23 | 1.25 | 1.0 | 2.2 | -1.2 |
| Calcite | 3.03 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Ankerite | 2.9 | 14.33 | 5.12 | 4.1 | 0.0 | 4.1 |
| Dolomite | 2.89 | 12.23 | 4.37 | 3.5 | 6.9 | -3.4 |
| Siderite | 2.8 | 3.15 | 1.13 | 0.9 | 0.0 | 0.9 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 4.5 | 1.22 | 1.0 | 0.0 | 1.0 |
| Witherite | 3.72 | 22.58 | 22.58 | 18.1 | 0.0 | 18.1 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| | | SUM | 124.90 | 100.0 | | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| DB 3 Month | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 78.4 | 69.1 | 9.3 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Pyroxene | 3 | 2.82 | 2.82 | 3.8 | 17.0 | -13.2 |
| Quartz | 3.34 | 0 | 0.00 | 0.0 | 3.4 | -3.4 |
| Clay | 4.5 | 0 | 0.00 | 0.0 | 0.7 | -0.7 |
| Calcite | 3.03 | 15.74 | 4.25 | 5.7 | 5.4 | 0.3 |
| Ankerite | 2.9 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 | 1.6 | -1.6 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 0.5 | -0.5 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0.0 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 | 2.3 | -2.3 |
| Witherite | 3.72 | 9.16 | 9.16 | 12.2 | 0.0 | 12.2 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0.3 | -0.3 |
| | | SUM | 75.06 | 100.0 | | |

| HBC 3 Month | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 70.5 | 71.9 | -1.4 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 10.2 | 10.20 | 12.2 | 16.4 | -4.2 |
| Quartz | 3.34 | 26.94 | 6.27 | 7.5 | 2.6 | 4.9 |
| Clay | 4.5 | 2.69 | 1.04 | 1.2 | 2.2 | -1.0 |
| Calcite | 3.03 | 2.62 | 0.71 | 0.8 | 0 | 0.8 |
| Ankerite | 2.9 | 15.42 | 5.51 | 6.6 | 0 | 6.6 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 | 6.9 | -6.9 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 3.39 | 0.92 | 1.1 | 0 | 1.1 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 83.46 | 100.0 | | |

| HBM 3 Month | | | | | | |
|--------------|---------------|-----------|----------------|---------------|------------------------|--------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) | Abundance in Unreacted | Total Change |
| Plagioclase | 3.19 | 100 | 58.82 | 69.4 | 64.1 | 5.3 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Pyroxene | 3 | 13.2 | 13.20 | 15.6 | 28.2 | -12.6 |
| Quartz | 3.34 | 8.62 | 2.00 | 2.4 | 1.4 | 1.0 |
| Clay | 4.5 | 1.55 | 0.60 | 0.7 | 0.6 | 0.1 |
| Calcite | 3.03 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Ankerite | 2.9 | 11.62 | 4.15 | 4.9 | 3.9 | 1.0 |
| Dolomite | 2.89 | 7.03 | 2.51 | 3.0 | 0 | 3.0 |
| Siderite | 2.8 | 3.5 | 1.25 | 1.5 | 1.8 | -0.3 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| Witherite | 3.72 | 2.24 | 2.24 | 2.6 | 0 | 2.6 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 | 0 | 0.0 |
| | | SUM | 84.78 | 100.0 | | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Data From Unreacted Samples

| DB Unreacted | | | | |
|--------------|---------------|-----------|----------------|---------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) |
| Plagioclase | 3.19 | 100 | 58.82 | 69.1 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 |
| Pyroxene | 3 | 14.43 | 14.43 | 17.0 |
| Quartz | 3.34 | 12.62 | 2.93 | 3.4 |
| Clay | 4.5 | 1.45 | 0.56 | 0.7 |
| Calcite | 3.03 | 17.04 | 4.61 | 5.4 |
| Ankerite | 2.9 | 0 | 0.00 | 0.0 |
| Dolomite | 2.89 | 3.88 | 1.39 | 1.6 |
| Siderite | 2.8 | 1.03 | 0.37 | 0.5 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 |
| Strontianite | 3.54 | 7.27 | 1.96 | 2.3 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 |
| | | SUM | 85.07 | 100.1 |

| HBC Unreacted | | | | |
|---------------|---------------|-----------|----------------|---------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) |
| Plagioclase | 3.19 | 100 | 58.82 | 71.9 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 |
| Pyroxene | 3 | 13.39 | 13.39 | 16.4 |
| Quartz | 3.34 | 9.22 | 2.14 | 2.6 |
| Clay | 4.5 | 4.61 | 1.78 | 2.2 |
| Calcite | 3.03 | 0 | 0.00 | 0.0 |
| Ankerite | 2.9 | 0 | 0.00 | 0.0 |
| Dolomite | 2.89 | 15.84 | 5.66 | 6.9 |
| Siderite | 2.8 | 0 | 0.00 | 0.0 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 |
| | | SUM | 81.79 | 100.0 |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

| HBM Unreacted | | | | |
|---------------|---------------|-----------|----------------|---------------|
| Mineral | Ref. Peak (Å) | Intensity | Intensity / WF | Abundance (%) |
| Plagioclase | 3.19 | 100 | 58.82 | 64.1 |
| Amphibole | 8.5 | 0 | 0.00 | 0.0 |
| Pyroxene | 3 | 25.89 | 25.89 | 28.2 |
| Quartz | 3.34 | 5.71 | 1.33 | 1.4 |
| Clay | 4.5 | 1.4 | 0.54 | 0.6 |
| Calcite | 3.03 | 0 | 0.00 | 0.0 |
| Ankerite | 2.9 | 10.01 | 3.58 | 3.9 |
| Dolomite | 2.89 | 0 | 0.00 | 0.0 |
| Siderite | 2.8 | 4.64 | 1.66 | 1.8 |
| Aragonite | 3.4 | 0 | 0.00 | 0.0 |
| Strontianite | 3.54 | 0 | 0.00 | 0.0 |
| Witherite | 3.72 | 0 | 0.00 | 0.0 |
| Vaterite | 3.29 | 0 | 0.00 | 0.0 |
| | | SUM | 91.81 | 100.0 |

Appendix B: X-Ray Fluorescence (XRF) Data

The following data is the entire major and trace element data gained from XRF analysis. The first column is the oxide, the second column states whether the data is as a weight percent (wt.%) or as parts per million (ppm). The last three columns are the actual XRF data for each sample.

| Oxide Symbol | | DB | HBC | HBM |
|--------------|-----|---------|---------|---------|
| SiO2 | wt% | 52.026 | 51.890 | 51.723 |
| TiO2 | wt% | 0.995 | 1.028 | 1.033 |
| AI2O3 | wt% | 13.787 | 13.955 | 13.802 |
| Fe2O3T | wt% | 13.029 | 14.055 | 14.027 |
| MnO | wt% | 0.235 | 0.237 | 0.244 |
| MgO | wt% | 5.368 | 5.931 | 5.729 |
| CaO | wt% | 8.635 | 10.207 | 10.229 |
| Na2O | wt% | 5.765 | 2.387 | 2.389 |
| К2О | wt% | 0.092 | 0.429 | 0.463 |
| P2O5 | wt% | 0.124 | 0.128 | 0.128 |
| Nb | ppm | 4.200 | 4.400 | 4.400 |
| Zr | ppm | 83.000 | 87.000 | 87.000 |
| Υ | ppm | 26.800 | 27.500 | 27.300 |
| Sr | ppm | 72.000 | 153.000 | 153.000 |
| U | ppm | 2.000 | 1.000 | 1.000 |
| Rb | ppm | 0.400 | 7.300 | 7.600 |
| Th | ppm | 3.000 | 2.000 | 2.000 |
| Pb | ppm | 6.000 | 4.000 | 5.000 |
| Ga | ppm | 17.000 | 18.000 | 18.000 |
| Zn | ppm | 84.000 | 106.000 | 103.000 |
| Ni | ppm | 27.000 | 28.000 | 28.000 |
| Cr | ppm | 24.000 | 28.000 | 26.000 |
| Ті | % | 0.810 | 1.020 | 1.010 |
| V | ppm | 276.000 | 314.000 | 314.000 |
| Ce | ppm | 19.000 | 19.000 | 22.000 |
| Ва | ppm | 26.000 | 97.000 | 83.000 |
| La | ppm | 9.000 | 7.000 | 9.000 |
| Fe | % | 0.106 | 0.105 | 0.104 |

Appendix C: SEM Spectra and Images

The following are SEM spectra analysis and images from Experiment 7, which was the 3 month experiment. Spectra and images are provided for each sample. First the spectra are shown, followed by the corresponding image. In each corresponding image is a small bright green cross mark indicating where on the image the spectrum represents.



DB 3month (Experiment 7)

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011





















HBC 3 Month (Experiment 7)




















HBM 3 Month (Experiment 7)



















Appendix D: SEM Elemental Mapping

The following images are elemental maps taken with the SEM. All of the elements being examines were the major cations that will react to form carbonate minerals. While using the SEM to locate and image carbonate minerals, sometimes the carbonates formed were so small (less than a micron) that it was extremely difficult to find them. Therefore, we utilized the below mapping technique to locate the major carbonate forming cations. Once we found these cations, we then focused our search in those areas. Where the color is brightest in the map is where the highest concentration of a given element is.



Elemental Maps from HBC Experiment 5

Map of Barium, shown if bright purple.



Map of Calcium, shown in red.



Map of Iron, shown in green.



Map of Magnesium, shown in yellow.



Map of Carbon, shown in blue.



Map of Strontium, shown in blue.



Maps of Various Elements from HBM Experiment 4

Map of Barium, shown in yellow.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011



Map of Carbon, shown in green.



Map of Iron, shown in purple.



Map of Magnesium, shown in red.



Map of Strontium, shown in gold.

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Appendix E: Grain Size Data

The following is all of the grain size data from each sample for experiments 1-7. Once samples were ground to a power in the shatter box they were then analyzed for mean grain size. The most important columns for grain size distribution are columns 1, 3 and 4. Column 1 identifies the size of the grain while the third column represents the percent of grains that did not fall through the sieve. The sieve sizes are given in column 2. The fourth column gives the percent of grains that passed through the sieve. To attain a cumulative grain size distribution curve you plot the first column verses the fourth column.

Deerfield Basalt

Experiments 1-6

| Size class | [mm] | retained [%] | passing [%] | SPHT3 | Symm3 | b/l3 | PDN |
|------------|-----------|--------------|-------------|-------|-------|-------|--------|
| 0,000 | 0,100 | 0,41 | 0,41 | 0,874 | 0,855 | 0,682 | 255657 |
| 0,100 | 0,125 | 0,42 | 0,83 | 0,828 | 0,847 | 0,654 | 63501 |
| 0,125 | 0,160 | 1,06 | 1,89 | 0,802 | 0,842 | 0,650 | 74558 |
| 0,160 | 0,200 | 2,00 | 3,89 | 0,769 | 0,834 | 0,640 | 68991 |
| 0,200 | 0,250 | 3,67 | 7,56 | 0,744 | 0,829 | 0,641 | 68400 |
| 0,250 | 0,315 | 6,33 | 13,89 | 0,707 | 0,818 | 0,637 | 56818 |
| 0,315 | 0,400 | 9,85 | 23,74 | 0,678 | 0,813 | 0,641 | 45033 |
| 0,400 | 0,500 | 12,68 | 36,42 | 0,658 | 0,814 | 0,645 | 28955 |
| 0,500 | 0,630 | 15,73 | 52,15 | 0,617 | 0,810 | 0,630 | 17982 |
| 0,630 | 0,800 | 15,58 | 67,73 | 0,583 | 0,801 | 0,617 | 8428 |
| 0,800 | 1,000 | 12,68 | 80,41 | 0,621 | 0,799 | 0,608 | 3572 |
| 1,000 | 1,250 | 8,31 | 88,72 | 0,583 | 0,771 | 0,611 | 1240 |
| 1,250 | 1,600 | 6,17 | 94,89 | 0,627 | 0,781 | 0,616 | 499 |
| 1,600 | 2,000 | 3,50 | 98,39 | 0,561 | 0,763 | 0,598 | 137 |
| 2,000 | 2,500 | 1,33 | 99,72 | 0,469 | 0,731 | 0,605 | 30 |
| 2,500 | 3,150 | 0,12 | 99,84 | 0,203 | 0,602 | 0,537 | 1 |
| 3,150 | 4,000 | 0,16 | 100,00 | 0,298 | 0,772 | 0,640 | 1 |
| 4,000 | 5,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 5,000 | 6,300 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 6,300 | 8,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 8,000 | 10,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 10,000 | 12,500 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 12,500 | 16,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 16,000 | 1,000,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |

Experiment 7

| Table of measurement results: | | | | | | | | |
|--|-----------|------------|------------|-------|-------|-------|--------|--|
| C:\Program Files\CAMSIZER\CAMDAT\NaughtonHMB\DB42_2mm007.rdf | | | | | | | | |
| Task file: SCP2_coarse.afg | | | | | | | | |
| | | | | | | | | |
| Size class | [mm] | retained [| passing [% | SPHT3 | Symm3 | b/I3 | PDN | |
| 0,000 | 0,100 | 1,35 | 1,35 | 0,867 | 0,851 | 0,673 | 159460 | |
| 0,100 | 0,125 | 0,82 | 2,17 | 0,803 | 0,835 | 0,638 | 21618 | |
| 0,125 | 0,160 | 1,28 | 3,45 | 0,771 | 0,828 | 0,634 | 15308 | |
| 0,160 | 0,200 | 1,74 | 5,19 | 0,715 | 0,807 | 0,608 | 10234 | |
| 0,200 | 0,250 | 2,12 | 7,31 | 0,695 | 0,804 | 0,616 | 6405 | |
| 0,250 | 0,315 | 2,90 | 10,21 | 0,610 | 0,781 | 0,551 | 4096 | |
| 0,315 | 0,400 | 4,21 | 14,42 | 0,653 | 0,806 | 0,563 | 2808 | |
| 0,400 | 0,500 | 7,88 | 22,30 | 0,742 | 0,849 | 0,617 | 2755 | |
| 0,500 | 0,630 | 10,81 | 33,11 | 0,741 | 0,857 | 0,612 | 1927 | |
| 0,630 | 0,800 | 11,97 | 45,08 | 0,752 | 0,854 | 0,660 | 1174 | |
| 0,800 | 1,000 | 10,45 | 55,53 | 0,806 | 0,868 | 0,653 | 485 | |
| 1,000 | 1,250 | 12,65 | 68,18 | 0,766 | 0,871 | 0,596 | 269 | |
| 1,250 | 1,600 | 13,56 | 81,74 | 0,823 | 0,870 | 0,658 | 169 | |
| 1,600 | 2,000 | 11,02 | 92,76 | 0,835 | 0,877 | 0,681 | 76 | |
| 2,000 | 2,500 | 6,80 | 99,56 | 0,817 | 0,863 | 0,660 | 20 | |
| 2,500 | 3,150 | 0,44 | 100,00 | 0,852 | 0,874 | 0,726 | 2 | |
| 3,150 | 4,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 4,000 | 5,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 5,000 | 6,300 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 6,300 | 8,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 8,000 | 10,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 10,000 | 12,500 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 12,500 | 16,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |
| 16,000 | 1,000,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 | |

MA Clean Energy Center CO2 Sequestration in Basalt 10/14/2011

Holyoke Basalt Massachusetts (HBM)

Experiments 1-6

| Size class | [mm] | retained [| passing [% | SPHT3 | Symm3 | b/I3 | PDN |
|------------|-----------|------------|------------|-------|-------|-------|--------|
| 0,000 | 0,100 | 0,89 | 0,89 | 0,881 | 0,858 | 0,676 | 329850 |
| 0,100 | 0,125 | 0,91 | 1,80 | 0,830 | 0,849 | 0,653 | 90230 |
| 0,125 | 0,160 | 2,19 | 3,99 | 0,801 | 0,842 | 0,640 | 98283 |
| 0,160 | 0,200 | 3,95 | 7,94 | 0,757 | 0,829 | 0,621 | 86354 |
| 0,200 | 0,250 | 6,70 | 14,64 | 0,734 | 0,824 | 0,625 | 78357 |
| 0,250 | 0,315 | 10,83 | 25,47 | 0,702 | 0,814 | 0,631 | 64649 |
| 0,315 | 0,400 | 15,18 | 40,65 | 0,672 | 0,808 | 0,632 | 45106 |
| 0,400 | 0,500 | 15,53 | 56,18 | 0,636 | 0,805 | 0,628 | 23063 |
| 0,500 | 0,630 | 15,85 | 72,03 | 0,613 | 0,799 | 0,623 | 12025 |
| 0,630 | 0,800 | 12,55 | 84,58 | 0,537 | 0,787 | 0,578 | 4385 |
| 0,800 | 1,000 | 7,37 | 91,95 | 0,590 | 0,779 | 0,604 | 1382 |
| 1,000 | 1,250 | 4,39 | 96,34 | 0,526 | 0,747 | 0,556 | 392 |
| 1,250 | 1,600 | 2,19 | 98,53 | 0,614 | 0,764 | 0,632 | 115 |
| 1,600 | 2,000 | 1,17 | 99,70 | 0,572 | 0,771 | 0,568 | 29 |
| 2,000 | 2,500 | 0,30 | 100,00 | 0,681 | 0,815 | 0,618 | 4 |
| 2,500 | 3,150 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 3,150 | 4,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 4,000 | 5,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 5,000 | 6,300 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 6,300 | 8,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 8,000 | 10,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 10,000 | 12,500 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 12,500 | 16,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 16,000 | 1,000,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |

Experiment 7

| Size class | [mm] | retained [| passing [% | SPHT3 | Symm3 | b/I3 | PDN |
|------------|-----------|------------|------------|-------|-------|-------|-------|
| 0,000 | 0,100 | 0,10 | 0,10 | 0,885 | 0,863 | 0,665 | 14511 |
| 0,100 | 0,125 | 0,02 | 0,12 | 0,865 | 0,874 | 0,648 | 605 |
| 0,125 | 0,160 | 0,02 | 0,14 | 0,808 | 0,847 | 0,608 | 285 |
| 0,160 | 0,200 | 0,05 | 0,19 | 0,785 | 0,887 | 0,566 | 218 |
| 0,200 | 0,250 | 0,08 | 0,27 | 0,713 | 0,862 | 0,521 | 181 |
| 0,250 | 0,315 | 0,33 | 0,60 | 0,680 | 0,865 | 0,468 | 341 |
| 0,315 | 0,400 | 0,86 | 1,46 | 0,701 | 0,870 | 0,480 | 431 |
| 0,400 | 0,500 | 2,86 | 4,32 | 0,730 | 0,860 | 0,555 | 888 |
| 0,500 | 0,630 | 4,70 | 9,02 | 0,751 | 0,869 | 0,587 | 807 |
| 0,630 | 0,800 | 7,62 | 16,64 | 0,677 | 0,845 | 0,520 | 573 |
| 0,800 | 1,000 | 8,63 | 25,27 | 0,797 | 0,866 | 0,629 | 369 |
| 1,000 | 1,250 | 14,40 | 39,67 | 0,773 | 0,869 | 0,605 | 332 |
| 1,250 | 1,600 | 19,89 | 59,56 | 0,809 | 0,865 | 0,640 | 244 |
| 1,600 | 2,000 | 20,56 | 80,12 | 0,820 | 0,871 | 0,664 | 131 |
| 2,000 | 2,500 | 16,60 | 96,72 | 0,819 | 0,871 | 0,694 | 59 |
| 2,500 | 3,150 | 3,28 | 100,00 | 0,770 | 0,846 | 0,671 | 8 |
| 3,150 | 4,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 4,000 | 5,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 5,000 | 6,300 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 6,300 | 8,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 8,000 | 10,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 10,000 | 12,500 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 12,500 | 16,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 16,000 | 1,000,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |

Holyoke Basalt Connecticut (HBC)

Experiments 1-6

| Size class | [mm] | retained [| passing [% | SPHT3 | Symm3 | b/I3 | PDN |
|------------|-----------|------------|------------|-------|-------|-------|--------|
| 0,000 | 0,100 | 0,42 | 0,42 | 0,877 | 0,857 | 0,682 | 233712 |
| 0,100 | 0,125 | 0,45 | 0,87 | 0,838 | 0,851 | 0,660 | 64201 |
| 0,125 | 0,160 | 1,16 | 2,03 | 0,808 | 0,844 | 0,653 | 77514 |
| 0,160 | 0,200 | 2,30 | 4,33 | 0,768 | 0,832 | 0,638 | 74692 |
| 0,200 | 0,250 | 4,72 | 9,05 | 0,749 | 0,830 | 0,646 | 83650 |
| 0,250 | 0,315 | 7,78 | 16,83 | 0,718 | 0,823 | 0,643 | 66809 |
| 0,315 | 0,400 | 11,64 | 28,47 | 0,683 | 0,815 | 0,646 | 50899 |
| 0,400 | 0,500 | 13,02 | 41,49 | 0,637 | 0,804 | 0,632 | 28091 |
| 0,500 | 0,630 | 14,10 | 55,59 | 0,601 | 0,798 | 0,622 | 15122 |
| 0,630 | 0,800 | 13,94 | 69,53 | 0,558 | 0,787 | 0,612 | 7064 |
| 0,800 | 1,000 | 10,53 | 80,06 | 0,612 | 0,786 | 0,610 | 2853 |
| 1,000 | 1,250 | 8,54 | 88,60 | 0,565 | 0,777 | 0,604 | 1202 |
| 1,250 | 1,600 | 6,35 | 94,95 | 0,637 | 0,781 | 0,621 | 512 |
| 1,600 | 2,000 | 3,21 | 98,16 | 0,625 | 0,790 | 0,625 | 128 |
| 2,000 | 2,500 | 1,49 | 99,65 | 0,478 | 0,734 | 0,607 | 30 |
| 2,500 | 3,150 | 0,23 | 99,88 | 0,416 | 0,688 | 0,646 | 3 |
| 3,150 | 4,000 | 0,12 | 100,00 | 0,371 | 0,708 | 0,686 | 1 |
| 4,000 | 5,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 5,000 | 6,300 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 6,300 | 8,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 8,000 | 10,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 10,000 | 12,500 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 12,500 | 16,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 16,000 | 1,000,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |

Experiment 7

| Size class | [mm] | retained [| passing [% | SPHT3 | Symm3 | b/I3 | PDN |
|------------|-----------|------------|------------|-------|-------|-------|------|
| 0,000 | 0,100 | 0,03 | 0,03 | 0,866 | 0,868 | 0,636 | 3434 |
| 0,100 | 0,125 | 0,01 | 0,04 | 0,856 | 0,877 | 0,639 | 463 |
| 0,125 | 0,160 | 0,02 | 0,06 | 0,833 | 0,872 | 0,615 | 361 |
| 0,160 | 0,200 | 0,04 | 0,10 | 0,770 | 0,834 | 0,575 | 207 |
| 0,200 | 0,250 | 0,19 | 0,29 | 0,594 | 0,854 | 0,366 | 434 |
| 0,250 | 0,315 | 0,52 | 0,81 | 0,635 | 0,852 | 0,410 | 621 |
| 0,315 | 0,400 | 1,62 | 2,43 | 0,730 | 0,866 | 0,526 | 1285 |
| 0,400 | 0,500 | 5,52 | 7,95 | 0,743 | 0,867 | 0,562 | 2284 |
| 0,500 | 0,630 | 7,56 | 15,51 | 0,750 | 0,864 | 0,600 | 1821 |
| 0,630 | 0,800 | 8,11 | 23,62 | 0,746 | 0,856 | 0,609 | 954 |
| 0,800 | 1,000 | 10,42 | 34,04 | 0,781 | 0,861 | 0,625 | 624 |
| 1,000 | 1,250 | 11,20 | 45,24 | 0,774 | 0,856 | 0,633 | 362 |
| 1,250 | 1,600 | 21,05 | 66,29 | 0,806 | 0,867 | 0,634 | 336 |
| 1,600 | 2,000 | 20,18 | 86,47 | 0,813 | 0,870 | 0,646 | 168 |
| 2,000 | 2,500 | 12,79 | 99,26 | 0,831 | 0,875 | 0,697 | 60 |
| 2,500 | 3,150 | 0,74 | 100,00 | 0,817 | 0,861 | 0,705 | 1 |
| 3,150 | 4,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 4,000 | 5,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 5,000 | 6,300 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 6,300 | 8,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 8,000 | 10,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 10,000 | 12,500 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 12,500 | 16,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |
| 16,000 | 1,000,000 | 0,00 | 100,00 | 1,000 | 0,000 | 0,000 | 0 |