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Review of CO₂ Storage in Basalts

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This report describes research sponsored by IEAGHG. This report was prepared by:

- Lydia Rycroft
- James Craig

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James Craig

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Further information or copies of the report can be obtained by contacting IEAGHG at:

IEAGHG, Pure Offices, Cheltenham Office Park Hatherley Lane, Cheltenham, GLOS., GL51 6SH, UK Tel: +44 (0)1242 802911 E-mail: mail@ieaghg.org Internet: www.ieaghg.org



REVIEW OF CO2 STORAGE IN BASALTS

Executive Summary

This technical review has been undertaken with the aim of providing a high level overview of the current status of basalts as an option for the geological storage of CO_2 . The review also includes a short section on the storage potential of ultramafic rocks.

Two high profile sites, CarbFix in Iceland and the Wallula project in Washington State have both injected and monitored CO₂ storage in basalts since 2012 and research has recently been published for both sites. Basalts are important storage sites to consider for CCS as they comprise approximately 10% of the Earth's surface and are often located in areas where no other storage options exist. Basalts have a high weight percentage of Ca, Mg and Fe rich minerals which react with CO₂ to form carbonates. At the pilot projects Wallula and CarbFix, in-situ carbonisation has been proven to occur and within much shorter timescales than initially predicted. In conventional deep sandstone aquifer storage sites, CO₂ remains buoyant for 1,000s to 100,000s of years and consequently this form of storage relies predominately on structural and solution trapping within the reservoirs to prevent CO₂ leakage.

Comparatively rapid mineral trapping created by reactive cations reduces the risk of leakage as the CO_2 becomes permanently trapped within the host reservoir rock's structure. The use of basalt would also increase the number of reservoirs available for storage as caprock integrity is only required for tens of years rather than thousands. This property could be beneficial for CO_2 storage by reducing the risks of leakage.

The depositional structure of basalts (as well as their mineralogy) is also beneficial for the storage of CO_2 . Basalts often form a series of layered deposits due to their volcanic origin. Basalt lava flows degas at the upper surface forming bubbles, 'vesicles', within the rock within a fine grained matrix. The presence of vesicles increases the porosity and permeability of the rock and the reactive surfaces. The top layer of basalt extrusions are often weathered altering the structure of the rock. Lava flows can also be interspersed with sedimentary layers. Weathering degradation of the rock, and impermeable strata, often leads to the formation of suitable caprocks. All of these factors increase suitability of basalt formations for use as storage of CO_2 .

Different technologies have been utilised for carbon sequestration in basalts compared to sedimentary formations. At the CarbFix experimental site CO_2 is dissolved into water and injected as an aqueous phase (i.e. not as pure CO_2) to promote rapid reactivity. Although sequestration at the site has been successful the quantity of water required may be a factor restricting large-scale CO_2 storage in basalts. Approximately 27 tons of water were required for every ton of CO_2 stored. CarbFix sourced this water from a local aquifer but for larger projects this may not be feasible. At the Big Sky pilot injection site in Wallula nearly a 1,000 tonnes of super critical CO_2 was injected in 2013. Borehole and pressure testing, plus surface sampling, confirmed that the CO_2 was retained in the target formation.

Overall, the two pilot sites conducted to date have shown great promise for the successful use of basalts as CO_2 storage reservoirs. The potential for further storage globally is vast, especially in areas such as India, the USA and possibly South Africa. Further research is still required, predominantly focusing on the amount of water required, where it can be sourced



from and at what expense. Further analysis is currently being undertaken at the U.S Wallula site as cores have only recently been extracted.

This review includes two examples of situations where basalt formations are in comparatively close proximity to large point sources of CO₂.

Ultramafic rock formations have high magnesium contents (>12% by weight) which are geochemically more reactive with CO_2 compared with mafic basalts. There is strong evidence of natural carbonation reactions in regions of the world where ultramafic formations are exposed, for example, the Oman ophiolite. However, ultramafic rocks are not as widespread in comparison with basalts and often form coarse grained plutonic complexes which have more restricted reactive surfaces. There are also occurrences of ultramafic lavas with limited global distribution compared with mafic basalts.

The use of mine tailings derived from ultramafic host formations has been investigated in South Africa as a means of CO_2 sequestration. The process could have application in Finland, Canada, Australia and the USA.



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1. Geological Storage of CO₂ in Basalts

1. Introduction

IEAGHG published a report in 2011 as an initial high level overview of the status of basalts as an option for the geological storage of CO_2 (IEAGHG, 2011). This 2016 technical review aims to provide an update to the 2011 report as injection has now started at two high profile pilot projects (Big Sky's Wallula project, US and CarbFix, Iceland). Recent data from both projects gives an insight into the reactions that are taking place and the timescales required for mineral formation. Field data and modelling has enabled the amount of CO_2 reacting with mafic elements to create stable carbonate minerals to be estimated.

Although sedimentary formations are considered to have the largest potential for CO_2 , basalts are present both on and offshore in large deposits in regions where suitable sedimentary formations do not exist. This study also reports on the new injection technologies used including the implications for the large volumes of water required in comparison to injection in conventional reservoirs. This review also includes a short section on the potential of ultramafic minerals to fix CO_2 by carbonation reactions.

2. Storage Concept

Basalts are dark-coloured basic, fine grained igneous rocks most commonly occurring in extrusive volcanic settings. They are defined by their mineralogy and grain size, comprising 45% to 85% (by volume) of mafic minerals with crystals under 2mm in diameter (fine grained) as shown in Figure 1. Mafic is a term used to describe a rock rich in magnesium and iron which includes basalts, comprised predominantly of plagioclase (NaAlSi₃O₈ – CaAl₂Si₂O₈) and pyroxene (XY(Si,Al)₂O₆ where X is predominately Ca²⁺, Mg²⁺, Fe²⁺, Na⁺ and Y is Fe³⁺, Al³⁺ and some other metal cations) but can include olivine (MgFeSiO₄). Ultramafic rocks (greater than 85% volume mafic minerals e.g. peridotite which is composed predominately of Mg-rich Olivine) also have storage potential because of their high magnesium content which makes it potentially more reactive than basalt, although it is not as extensive and tends not to form permeable lava flows.



Figure 1: Igneous rock classification by mineral composition (from M.Y Farooqui et al.,) 2009. Basalt classification is highlighted in red box.



One of the main advantages of storing CO_2 in basalts are the geochemical reactions that take place. These reactions occur naturally (they are exothermic) allowing the mineral trapping of CO_2 to occur at a faster rate compared with conventional sedimentary aquifers. The reactivity of CO_2 can be enhanced by dissolution when the gas is dissolved in water and then injected into the target formation. In these circumstances dissolution reactions are fast compared with silicate minerals with higher Si:O ratios such as Na and K-rich-feldspars and quartz. Mafic and ultramafic rocks release divalent metal cation such as Ca^{2+} at relatively rapid rates, consequently in-situ mineralisation occurs over a comparatively short timescale.

Basaltic formations contain high levels of calcium, magnesium and iron which react with the brackish water (pH of 8.5 to 9.2 commonly found in the formations) to form carbonate minerals due to CO₂ injection. This occurs as the injection of CO₂ alters the pH to 3.5 or lower (McGrail et al., 2003) and causes iron rich phases to become unstable and dissolve (e.g. pyroxene and olivine). These then react with CO₂ to form carbonate minerals, most commonly calcite (CaCO₃), dolomite (Ca_{0.5}Mg_{0.5}CO₃), magnesite (MgCO₃) and siderite (FeCO₃). As highlighted in the IEAGHG 2011 report, laboratory experiments have been conducted to predict these reactions and as Gislason et al (2010) described the most significant reactions expected are:

- 1. $CO_{2(g)} + H_2O = H_2CO_3$
- 2. $H_2CO_3 = HCO_3^- + H^+$
- 3. (Fe, Ca, Mg)²⁺ + CO₂ + H₂O = (Fe, Ca, Mg)CO₃ + H⁺
- 4. $Mg_2SiO_4 + 4H^+ \rightarrow 2Mg^{2+} + 2H_2O + SiO_{2(aq)}$
- 5. $CaAl_2Si_2O_8 + 8H^+ \rightarrow Ca^{2+} + 2Al^{3+} + 2SiO_{2(aq)} + 4H_2O$

The first two equations describe CO_2 dissolution in water. Equation 3 describes metal cations released into the formation water in basalts leading to carbonate formation with the release of H⁺ ions. Reactions 4 and 5 show the effects of acidification on silicate minerals releasing the cations needed to form carbonates (IEAGHG, 2011).

A series of models and high pressure experiments of CO₂-basalt interactions were undertaken by R. J Rosenbauer et al., 2012. The 'CHILLER' modelling program was used to simulate reaction paths and predicted an equilibrium alteration assemblage of calcite, magnesite and siderite. The only secondary carbonate identified in the simulations was ferroan magnesite. The overall reaction extent in the simulations was controlled by bulk magnesium content in the model, with an 8% Mg content being equivalent to 2.6 x 10⁸ metric tonnes (Mt) of CO₂/km³. High pressure experiments showed that basalt had a high reactivity with CO₂ acidified brine, the maximum extent and rate of reactions occurred at 100°C, 300 bar (which are common reservoir conditions at approximately 3,000m depth). Modelling conducted by Rosenbauer has revealed the implication of the volume increase noted for some reactions. For example the transition of olivine to form magnesite ($Mg_2SiO_4 + 2H_2CO_3 = 2MgCO_3 + 2H_2O + SiO_2$) leads to a volume increase. This is significant as it may block pores and inhibit the injectability of the reservoir in deep Mg-rich basalts. This is currently a hypothesis and requires further research. Computer simulations have also been undertaken to model the injection of CO₂ with H₂S impurities into basalt formations (Bacon et al., 2013). The model results predicted that CO₂ is trapped as calcite within decades and H₂S is trapped as pyrite within a few months.

The structural deposition of basalt is also beneficial for storage as its vesicular texture increases porosity and permeability. Basalt forms in layered extrusive deposits and often becomes more vesicular towards the top of deposits. Vesicles lead to macro-porosity but the permeability of basalts can be highly variable dependant on cooling rate, number of interflow zones and the thickness of the flow. The large-scale and episodic eruptions of basalts over geological time-



scales also means there is a vast worldwide distribution, as shown in Figure 2. Most large deposits are 'flood basalts' which form large volcanic provinces, plateaus and mountain ranges. The most famous occurrences are the Deccan traps in India (with a surface area over 1,500km²), the Siberian Traps in Russia (700km² surface area) and the Columbia River basalts of the northwestern United States. Basalts most commonly form at mid-ocean ridges and hence comprise most of the ocean floor and approximately 10% of the total continental surface area. In exceptional circumstances, for example Iceland, subaerial accumulations can occur at spreading centres.

Igneous deposits often vary in mineralogy between successive lava flows and so can alternate between rock-types, e.g. mafic and ultramafic. Ultramafic rocks contain over 85 wt% mafic minerals and also have a high potential for mineral carbonisation (see Section 3). Laboratory experiments have shown olivine to react to form carbonates hundreds to thousands of times faster than other silica saturated rock forming minerals.

As storage in basalts is strongly controlled by the chemical reactions that take place, the impurities within the CO_2 injected can have a significant impact. Laboratory experiments have been conducted to analyse the effects for CO_2 -H₂S-H₂O interactions (Schaef et al., 2013) and showed the formation of pyrite (FeS₂) prior to any carbonation reactions. The amount precipitated increased with Fe content and the amount of H₂S injected. The purification of CO_2 during the capture process is the most expensive part of a CCS project. At CarbFix H₂S was injected with the CO_2 to reduce cost and to study the impact.

Novel injection techniques have been developed with the aim of increasing solubility trapping of CO_2 storage in basalt reservoirs. Rather than pure supercritical CO_2 being injected, the CO_2 is dissolved in water to allow for in-situ carbonisation reactions to occur quickly. This technique was demonstrated at CarbFix but required large volumes of water, approximately 27 tons of water for every ton of CO_2 (Oelkers et al., 2008). Further details on the techniques used are included in the complete project descriptions in Section 3.

The rock demand for mineral sequestration is large. 5.9 tons of forsterite (Mg-olivine) or 8.8 tons of basaltic glass are required to fix one ton of CO_2 assuming complete dissolution of the rock and complete conversion of all the divalent cations to carbonates. One of the major challenges in mineral sequestration of CO_2 is to maximise the fraction of divalent cations that precipitate as carbonates compared with other secondary minerals such as oxides, clays and zeolites which have the potential to reduce permeability.

Dissolution rates are influenced by the crystallinity and rock composition. Volcanic glass can release Ca^{2+} about twice as fast as a fully crystalline rock of the same composition. Basaltic glass dissolution rates increase with increasing pH ie alkaline conditions. The dissolution rate of forsterite decreases with increasing pH. Decreasing pH from ~10 to 3.5 will increase dissolution rates by about two orders of magnitude but will have a negligible effect on basaltic glass dissolution rates.

The temperature and injection fluid composition can be varied to optimize divalent cation release rates. However, temperature in combination with pH can influence dissolution and the solubility of CO_2 in water decreases. Increasing temperature from 0 to 100°C at pH 9 results in a 4.5 order of magnitude increase in dissolution rate for aluminium but at pH 3.5 the rate only increases by a factor of 60. At high temperature the porosity declines and therefore the reactive surface area tends to decrease.



Injection systems therefore need to be fine-tuned with respect to reactive surface areas, rate of injection and the partial pressure of CO_2 to avoid excessive precipitation and clogging. The mineral-fluid interfacial surface area can be maximised by targeting porous rock formations or causing hydro fracturing during CO_2 injection. As reaction progresses secondary mineral formation could block the reactive surface slowing the dissolution process and reducing permeability.





Figure 2 Global distribution of Flood basalts



3. Ultra Mafic Rock Storage Potential

Other igneous rock formations that have high Mg contents also have the potential to trap atmospheric CO₂ by carbonation reactions. Ultramafic rocks commonly occur in the mantle as peridotite. This is a dense coarse grained rock often associated with ophiolites (obducted sections of oceanic crust) but also as massifs within mountain fold belts. These occurrences are comparatively rare compared with the distribution of flood basalts. In the geological past, predominantly in the Archean (4.0 - 2.5B years ago), ultramafic volcanic rocks known as Komatiites were extruded with very high Mg contents of >18% by weight. Despite this desirable property Komatiites are also rare although they are found in South Africa, Zimbabwe, Canada, the USA and Australia. Ultramafic rocks are therefore less common than basalts at the Earth's surface but are host to important metal deposits such as Cr-PGE (Lamont-Doherty Earth Observatory presentation 2013). The distribution of ultramafic rock formations of different origins has been complied by Columbia University and the US Geological Survey as a potential CO₂ sequestration resource (Krevor, S.C. et al, 2009).

Peridotite can consist of over 90% olivine, the most reactive silicate mineral, which forms magnesite through the following reaction:

 Mg_2SiO_4 (olivine) + 2CO₂ (from gas or fluid) = 2MgCO₃ (magnesite) + SiO₂ (quartz)

Ultramafic rocks, particularly peridotite which has Mg contents of >12% by weight, can be affected by dissolution and carbonation processes. Carbonation occurs when Ca^{2+} ions react with bicarbonate to form calcite. Mg rich minerals including Forsterite react with CO₂ in the presence of water to form hydrated carbonates and silica.

 $Ca^{2+} + HCO_3^- \rightarrow CaCO_3 + H^+$

 $MgSiO_4 + 2CO_2 + 2H_2O \rightarrow MgCO_3$ (hydrated) + SiO₂ (not balanced)

 CO_2 can also lead to the serpentinisation of peridotite and the formation of carbonate minerals such as calcite and magnesite.

 $Mg_2SiO_4 + CaMgSi_2O_6 + 2CO_2 + 2H_2O \rightarrow Mg_3Si_2O_5(OH)_4 + CaCO_3 + MgCO_3$

Mg-Olivine + CaMg-Pyroxene \rightarrow Serpentine + Calcite + Magnesite

These natural processes can lead to substantiate mineralisation and deposition of carbonates in regions of the world where there are sub-aerial exposures of ultramafic igenous formations for example the Oman ophiolite.





Figure 3 Natural carbonation leading to carbonate mineral precipitation in the Oman Ophiolite ultramafic exposed peridotite formations (Paukert and Matter, 2013).

Estimates of the volume of carbonate deposits formed during weathering of peridotite could be in the region of 40,000 tonnes/year within the Samail ophiolite equivalent to \sim 2 tonnes/km³ of peridotite (Kelemen and Matter, 2008). Although carbonation reactions are relatively rapid they are constrained by fracture density within the formation.

The relatively rapid fixation of CO_2 by Mg-rich minerals could also be exploited by using mine tailings with high concentrations of Mg-rich minerals. The concept has been proposed by South Africa which produces large quantities of crushed ultamafic material from Platinum group mineral mines. A study conducted in 2011 esimated 13.9 M tonnes of CO_2 could be sequested annually using this process (Doucet, F.T. 2011). The report has matched large point sources of CO_2 with ore deposits related to ultramafic rocks. This potential technological solution could also be of interest in Finland, USA and Canada.

4. Status of Pilot Field Demonstrations

4.1 Carb Fix Pilot Site, Iceland

The CarbFix pilot project in Iceland has undertaken two phases of CO_2 injection into basaltic formations beginning in 2012. The pilot project sources CO_2 from the Hellisheidi geothermal power plant (south-east of Reykjavik) where a gas separation station has been developed alongside full pipeline infrastructure for the transportation and injection of CO_2 . The objective of the project was to demonstrate CO_2 storage from the formation of environmentally benign carbonate mineralisation. The first phase of project injected 175 tons of pure CO_2 from January to March 2012 followed by 73 tons of CO_2 -H₂S in phase II from June to August 2012. Hydrogen sulphide (H₂S) was included in the testing as it is a major constituent of geothermal gases at this site. Injecting a mixture of gases would dramatically decrease the cost of capture as the separation process would require purification.

The site has a 2,000m deep injection well (HN02) and eight monitoring wells from 150 to 1,300m depth (Figure 4). The target storage formation is at 400-800m deep and consists of



basaltic lavas and hyaloclasites (a breccia rich in volcanic glass created by eruption under water or ice or by sudden quenching by water). As the formation is shallow, a novel injection technique was used where CO_2 was co-injected with water as an aqueous phase into the reservoir. This prevents the CO_2 forming a buoyant plume and allows instant reactions with Ca-Mg-Fe rich reservoir rocks. The water and CO_2 were injected via separate pipes (HN-2) to 360m depth where the CO_2 is then released via sparger into the water stream. This mixture was then carried to 560m where it was released into the reservoir. Mass balance equations show that the technique should allow for solubility trapping to occur within 5 minutes of injection into the storage reservoir (Sigfusson et al. 2015). Injection rates were on average approximately 70 g/s for CO_2 and 1,800g/s for water. In total 5,000t of water have been injected and 175t of CO_2 in the two phases of injection. The reservoir pH was 8.4 - 9.4 at a temperature range from 20-30°C. Tracers were injected to track the fate of the CO_2 as conventional seismic monitoring techniques could not be used. Full reservoir characteristics including geochemical properties are reported by Alfredsson et al., 2013.

A CarbFix report summary (European Commission, 2015) has concluded that the site has successfully demonstrated solubility trapping of CO_2 which occurred immediately and that CO_2 was trapped within carbonate minerals in a few years. The injectate was first detected using SF₆ tracer 56 days from the start of injection at a monitoring well. Differences between the predicted dissolved inorganic carbon and ¹⁴C content measured indicate that more than 95% of the injected CO₂ was mineralised within two years between the two monitoring wells.

The design of the new injection technology was led by Orkuveita Reykjavikur (OR), an Icelandic utilities company, with participation from the Centre national de la recherche scientifique (CNRS, National Centre of Research in France) and the University of Iceland (UI). A gas separator system was designed and optimised to generate gas for injection at the site sourced from the Hellisheidi power plant. The CO_2 is from a natural magmatic source derived from a geothermal power plant. The injection technology was designed to promote the dissolution of the injected gas as an aqueous phase to reduce the buoyancy of the CO_2 and promote rapid carbonate mineralisation. A detailed description of the injection system is shown in Figure 5. This in turn means caprock integrity is less important as there is a lower risk of atmospheric leakage.





Figure 4 Stratigraphic section of the CarbFix pilot injection site (modified from Alfredsson et al, 2008)

The target injection formation is present over a depth interval of 400-800m and consists of basaltic lavas and hyloclastite with calcite and Ca-zeolites as common alteration minerals. The temperature at injection depths is 30 - 50°C. The composition of the gas varies with time. CO_2 concentrations range from 60 to >100 mmol/kg. This gas stream also contains H₂S, H₂ and N₂.

Current estimates based on research experience to date suggest 100 kg CO_2/m^2 could be stored over a 500m - 1,000m depth interval across Iceland's high-temperature geothermal zone within the central rift. If this estimate is extrapolated over 1% of the island's land area, equivalent to 1,000 km², then 50 Gt of CO_2 could be potentially stored. One potential drawback with high reactivity, and associated mineralisation, is a reduction in permeability caused by precipitation leading to clogged pores within the host basalt.





Figure 5: Design of the carbon-dioxide injection system. The natural water level in well is 80–90 m below the surface. (1) CO₂ injection pipe, (2) water injection pipe, (3) CO₂ sparger located at a depth of 330–360 m, (4) outer mixing pipe extending to a depth of 540 m, (5) mixer at 420 m, (6) fluid outlet at 540 m, (7) service pipe for downhole sampling and observations, (8) service entry to well head space. (Siguffson et al., 2015)

Storage costs for the pilot programme at CarbFix have been estimated to be $344 \notin tCO_2$ (NPV 0). If a full-scale CO₂ storage project at the Hellisheidi site (ie assuming all the CO₂ is sourced from the Hellisheidi geothermal power output) was progressed costs could fall to $25.8 \notin tCO_2$ (15% IRR) - $31 \notin tCO_2$ (20% IRR). At $22.5 \notin tCO_2$ NPV is 0 (i.e. no profit). Further reductions $10.5 \notin tCO_2$ (15% IRR) – $12.3 \notin tCO_2$ (20% IRR), break-even price is $9.3 \notin tCO_2$ (NPV 0) might be achieved with a Maximum Reservoir Exploitation (MRE) level of development (Ragnheidardottira, E. et al, 2011). The MRE level is over a 30 fold decrease in cost compared with the cost for the pilot plant.



4.2 Big Sky

The Wallula project in Washington State, partially funded by the Big Sky Regional Partnership program, is a pilot site injecting CO₂ into the Grande Ronde basalt formation. This is the world's first pilot stage super critical CO₂ injection into a basalt formation. The formation is part of the approximately 200km^2 Miocene Columbia River Basalt Group, a flood basalt sequence with over 300 individual flows and an estimated volume of $200,000 \text{ km}^3$. Conservative estimates of CO₂ storage capacity range from 10 - 50 Gt (McGrail et al., 2006). The CO₂ was of food-grade standard sourced from refineries in Washington State or California. An injection well was drilled to approximately 1,250m as modelling suggested this was the optimum depth for sequestration (http://www.bigskyco2.org/basalt_injection, viewed 12.08.2016). Nearly 1,000 tons of CO₂ was injected into the basalt formation from July to August 2013 at a rate of 40 Mt/day. Three brecciated zones between 828 - 887m were selected for the injection programme.

A baseline seismic survey was successfully completed in 2007, the first (known) survey to image basalt with a thin sediment cover (Sullivan et al., 2011). This site characterisation showed no faults were present at the site and the basalt deposits were shown to be suitably thick. Further structural analysis of the geology of the site was made from the drilling of the injection well, as shown in Figure 6.



Figure 6: (A) Schematic illustration of typical flood-basalt flow. The vertical dimension ranges from 101to 102m. (B) Generalized illustration of borehole geology from the injection well drilled at the Walulla Basalt Sequestration Pilot Project. The composite injection zone comprises theSlack Canyon #2 flow top (829–844 m bgs), Slack Canyon #1 flow top (853–859 m bgs), and the Ortley flow top (865–874 m bgs). (R.M. Pollyea, 2016).

The figure illustrates the variable nature of the basalt which has a direct impact on permeability. The effect of this relative permeability on injection pressure was studied via modelling experiments (R.M. Pollyea, 2016). A simulation of the Slack Canyon ~2m flow top deposit showed that a >50MPa variability in injection pressure was possible with varying relative



permeability. This investigation demonstrates the importance of pre-injection site characterisation in determining the structural deposition of the basalt and its petro-physical properties.

Pre-injection hydrologic analysis has identified a 50m thick injection zone with three brecciated interflow zones separated by dense interior flows at the Wallula site (McGrail et al. GHGT10 2011). Further extensive hydraulic pumping tests were then conducted in 2012 to further analyse the conductivity of the shallower basalts. Resolving the hydraulic conductivity of the rocks led to the issue of a State permit for an injection with a 1,000Mt limit. In total, 977 tonnes were injected to make sure the permit was not exceeded. Tracers were added to the CO₂ stream to help scientists successfully observe and examine movements within the basalt reservoir. Extended hydrologic well tests conducted during late 2012 indicated no evidence of vertical migration. Soil gas monitoring and water sampling from monitoring wells, plus residual saturation tool logging, showed that CO₂ was only present in the target reservoir.

Combined with groundwater chemistry and geophysical wellbore surveys, the site was the first to provide field evidence of carbonate mineral formation within a basalt formation. Prior to site closure, 50 core samples were taken from the injection interval in June 2015 for geochemical and physical analysis. Two mineral phases were found in 1mm nodule samples and as cement in the vesicles (McGrail et al., 2016). The carbonate nodules were identified as ankerite (Ca(Fe,Mg,Mn)(CO₃)₂) whereas unreacted samples formed calcite. Further conclusive evidence of in-situ carbonisation was provided through isotropic analysis. The ankerite had depleted δ^{13} C compared to the natural carbonates which indicates they formed from the injected CO₂. The combination of data sources showed that carbonate minerals were formed within 24 months of emplacement, permanently storing the CO₂. Geophysical data collected before and after injection showed up to 90% CO₂ saturation in the upper flow-top interval.

5. Technical Challenges

The two case studies currently being undertaken in Iceland and Washington State have shown that carbon mineralisation can occur within years to decades in basalt formations. The CarbFix site exceeded modelled expectations with over 95% of injected CO_2 forming minerals within two years of injection. Storage at the Wallula project has shown mineral formation within a year, but researchers have not yet to made in depth quantifications on the amount of mineralisation that has occurred. Most models analysed the amount of CO_2 that would enter aqueous phase but further geochemical modelling and laboratory work is required to estimate the rate of mineral precipitation. Experiments conducted by Rosenbauer et al., 2012 predicted that the maximum precipitation rates occur at 100°C and 300 bar. The uptake at this optimum condition ranged from 8% by weight for a typical tholeite (silica saturated and low alkali basalts) to 26% for a picrite (Magnesium rich olivine-basalt) which is coincident with the magnesium content of the rock. At the CarbFix site these predictions were exceeded with 85% of mineral capture occurring within 5 years. For an average basalt with a MgO content of 8% an estimated 2.6 x 10^8 tonnes of CO_2 could be trapped per km³ basalt.

As studied by Rosenbauer et al., 2012 in laboratory experiments, the volume changes that occur during carbonate mineral formation may lead to a reduction in pore space. At CarbFix there were no unexpected increases in pressure detected in the reservoir (European Commission, 2016) although an initial sudden increase did occur due to the sparger becoming clogged up. The sparger is part of the injection equipment and controls bubble size and hence dissolution rates. The sparger was positioned at the end of the gas pipe and therefore acted as a filter which



led to mineral build up and a blockage forming. This problem was addressed by increasing the hole size from 0.01mm to 1mm.

Extensive modelling was undertaken for the Wallula Project to establish the permeability distribution in the reservoir and hence potential injection rates. Low injection rates were maintained to prevent over-pressurisation and pressure recovery data showed a transition to a lower permeability zone approximately 50m from the well which had not been expected (McGail et al., 2014). No increases in pressure due to decreases in porosity or permeability were experienced at either pilot site.

The potential degradation of the caprock within basalt formations was also analysed in a variety of studies. The CarbFix project reported on caprock reactivity to further improve geochemical modelling and assess levels of integrity. At CarbFix, diopside (a common basaltic mineral) was shown to be unaffected by calcite precipitation (European Commission, 2016). This implies the integrity of the caprock should not be influenced by the injection of CO_2 but further long-term analysis will be required. Prior to well decommissioning in June 2015 over 50 sidewall cores were extracted from the injection interval for geochemical and geophysical evaluation. Overall in both projects basalt proved to be successful in retaining CO_2 and due to the fast carbonate mineral precipitation rate buoyant CO_2 will only have to be trapped for very short timescales.

The CarbFix site implemented a novel technique for the injection of CO_2 incorporating the stream into water to form an aqueous phase prior to injection. This technique proved to be successful as it allowed reactions with the basalt host rock to occur quickly and prevented a buoyant reservoir of CO_2 forming. This required a large volume of water which may prove problematic for future commercial-scale projects. On average 27 tons of water were required for every ton of CO_2 stored (Oelkers et al., 2008). This was manageable for the CarbFix site as water was sourced from the local groundwater. It has been hypothesised that seawater could also be used which would prove less problematic for offshore projects but geochemical reactions may be influenced. At the Wallula site pure supercritical CO_2 was injected (as is common at most CCS sites) and although no precipitation quantifications have been made the technique also proves to have been successful in storing CO_2 although a buoyant reservoir may be present for longer time periods.

As CO₂ storage in basaltic formations is driven by chemical reactions temperature plays a key role. At the CarbFix site there is a steep geothermal gradient of 170° C/km (Alfredson et al., 2013) but the injection formation is at a shallow depth of 400-800m. The formation waters at this depth have a temperature of 18-33°C (Sigfusson et al., 2015) and a pH of 8.4 to 9.4. Laboratory experiments and modelling conducted by Rosenbauer et al., 2012 concluded that CO₂ is taken up from solution at all temperatures from 50 to 200 °C, 300 bar but the maximum extent and rate of reaction occurs at 100°C, 300 bar.

In the European Commission's summary report on the CarbFix project, reducing monitoring costs was highlighted as a major benefit of using basalt formations for CCS. Once the CO₂ has been precipitated as carbonate minerals, the risk of leakage is greatly reduced and hence less monitoring is undertaken. If this occurs within 5 years, as shown by the two pilot sites, this could greatly reduce the amount of monitoring required compared to conventional CCS sites and hence reduce project costs. At CarbFix the estimated costs of CO₂ transport were 17 \notin ton (Ragnheidardottir et al., 2011) which were calculated pre-injection. It was concluded that costs would be dependent on CO₂ flow rates as low rates increased the relative capital cost whereas electricity and water costs became more significant at higher flow rates. Costs published on



the CarbFix website (https://www.or.is/en/projects/carbfix/faqs, August 2016) estimate that given a 30 year lifetime for equipment the costs are \$30/ton (for capturing, transporting and injecting). This is significantly lower than the Global CCS Institute's generic estimate of \$60-130/ton for conventional methods. The low costs are associated with the capture method where H_2S was not separated and hence costs were reduced. No data relating to costs of the Wallula Project in Washington State has been made publically available.

6. Future Potential Sites

The key properties required for a potential basalt CCS site are: suitable structural deposit (caprock, porosity and permeability); general proximity of basalt formations to CO_2 sources; water availability for CO_2 injection if the CarbFix method is implemented; and a suitable location to allow for infrastructure requirements and social acceptance of the project. Countries with a large potential for CO_2 storage in basalts (other than Iceland and the USA) are addressed below.

6.1 Australia

The Antrim Plateau volcanic province in Northern Australia covers an area of flood basalt deposits in excess of 12,000km². Despite the scale of this igneous province its geographical isolation would not make it a preferred choice for CO₂ storage.

There has been more recent (within the last two million years) small-scale volcanic activity across Victoria. Extensive basalt flows have formed a thin veneer, generally less than 50m (Earthresources Victoria Government website 2016). The limited thickness of basalt lava flows suggests that this region would not be suitable for CO_2 storage

6.2 India

The Deccan Traps in India have a great potential for CCS as they consist of 512,000 km³ of basalt deposits. These formations are located on the Deccan plateau in west-central India. A review of storage potential in India was conducted by IEAGHG, 2008. Although basalts were not considered in the report as viable options for storage at the time (technology advancements for basalt storage were not well developed in 2008), their distributions were noted. A good correlation was noted between basalt formation locations and many large point sources, as illustrated in Figure 7.



Figure 7: Map illustration showing basalt storage potential in relation to CO₂ sources. Pinked hashed boxes show basalt locations and black circles show CO₂ source (IEAGHG, 2008).



The Deccan Traps have characteristic formation features that are common to other flood basalts. Field exposures from the Toranmal Section of the northern Deccan Traps, have major vesicular-amygdular zones which mark the top of flows (Mahoney, J.J et al, 2000). Thicknesses of 10-120m have been reported, although compound flows of between 450 m and 710m have also been recorded. These flows exhibit large cooling colonnades with middle entablatures. If these features extend laterally they would help to enhance injected CO_2 .



Figure 8. Simplified geological map of India. \Diamond = Rajmahal Traps

There is another much smaller basaltic province, the Rajmahal Traps, in eastern India within the state of Jharkhand with the potential for CO_2 storage (Figure 8). The deposits are up to 608m thick (at Rajmahal hill) with a shallow 2 to 5° dip. Although this formation extends over a much smaller area compared with the Deccan Traps it is within 30 km of two large and relatively modern coal-fired power stations (Kahalgaon 2,340 MW and Farakka 2,100 MW) (Wikipedia websites on Indian power planst, 2016). The proximity of these power stations to the Rajmahal is evident from the satellite image depicted in Figures 9 and 10. The Traps form a series of hills trending approximately north-south between the two power station locations and can be distinguished from the dark green shading in the image and by comparison with the simplified geological map of the Rajmahal Traps to the left (Geological Survey of India, 1954-55).





_____|20 km

Figure 9. Geological map of the Rajmahal Traps (left, Source: Geological Survey of India, 1954-55) and their topographical expression (right, Google maps), Jharkhand State India. The red markers denote the locations of the Kahalgaon and Farakka coal-fired power stations



Figure 10. Location of the Kahalgaon and Farakka coal-fired power stations in eastern India, Google maps.

Originally basalt storage was not considered a mature concept and hence storage estimations were not made. Now that storage has been shown to be viable in two separate projects basalt



formations in India require further in depth analysis as a viable storage option. The proximity of two comparatively modern (post 1986) coal-fired power stations to a large basalt formation suggests storage potential is worth further investigation.

6.3 South Africa

South Africa is heavily dependent on coal-fired power stations that use coal mined from seams within the Karoo Supergroup which occupies a basin in the centre of the country. However, the most promising CO₂ storage sites are located hundreds of kilometres away within the coastal Mesozoic Basins and offshore oil and gas fields that fringe the continent (Council for Geoscience South Africa, 2010). The challenge presented by the Karoo Supergroup is the prevalence of low porosity / permeable sediments including sandstones. The Karoo Supergroup is also heavily intruded by dolerite which has the effect of compartmentalising successions and potentially limiting lateral CO₂ storage sites. The dilemma posed by the distance between large quantities of carbon emissions from point sources and suitable CO₂ storage sites might be partially met from storage in basalt formations. The largest flood basalt in the region is the Karoo-Ferrar deposit covering more than 140,000km² of southern Africa (Figure 11). These formations have been described as basaltic lavas that began with the eruption of subaerial and locally subaqueous laves, initially a few metres to tens of meters thick (Ross, P.-S et al, 2005). There were a series of non-explosive to explosive eruptions of basaltic magma caused by its interaction with surface water or groundwater. Lava flows and interspersed with thinner ash beds. Above these deposits are basaltic tuffs and tuff-breccia. The later volcanoclastic deposits also include basalt- andesitic compositions (see Figure 11). The fine grained nature of these formations suggests that they would be relatively reactive with CO₂, although lavas with an andesitic composition would be less reactive compared to basalt because of their higher silica content and lower magnesium content. The distribution of basaltic lavas in South Africa would favour those formations in closest proximity to the main coal-fired power stations to the east of Johannesburg (see Figure 12). There are Karoo basalts between 110 and 150 km north of the city in the Sprinbok Flats area of the country which can reach a thickness of 600m (Visser, J.N.J., 1984). These basalt formations are approximately 100-130 km from large coal-fired power stations, for example Duhva (3,600 MW) and Kusile (4,800 MW under construction) (Wikipedia website on South African power plants). There are two power stations in Limpopo Province, Matimba (3990 MW) and Medupi (4,764 MW under development), which are in very close proximity to another basalt formation to the northwest near the border with Botswana (Figure 12). However, basalts at this location are much thinner, reaching up to 75m, and may be less suitable as storage candidates. There are extensive basalt outcrops further south and across Lesotho but these might not be accessible for CO₂ storage and are more remote from the large point sources of CO₂ east of Johannesburg.





Figure 11. Igneous intrusions in the Karoo Basin, South Africa Svensen, H., et al., 2007.



0 50 100 150 200 Kilometr

Figure 12. Geological map of northern South Africa (Council for Geoscience, 2003) with locations of three reference coal-fired power stations \Diamond . The Karoo basalt formations are depicted in dark purple.

Intermediate and plutonic equivalents of basalt, dolerite and gabbro, are widely distributed throughout the Karoo basin but they are less suitable because they lack porosity and fine gain size.

Another factor which might constrain CO_2 storage in basaltic formations in South Africa is the availability of water. South Africa faces a growing demand for water estimated to be 17.7Bm³ by 2030, but supply may only reach 15Bm³ by that date. There are also competing demands



for water as the country's economy expands particularly from agriculture, power generation and mining.

7. Conclusions

Since IEAGHG's review on storage in basaltic formations conducted in 2011, two pilot sites have shown the technique to be feasible with safe and efficient injection demonstrated. The porosity and permeability of the formations were found to be suitable with no pressure anomalies encountered (within the formations) and the chemical composition has proved to be beneficial to the mineral trapping of CO_2 on very short timescales.

The CarbFix Project has demonstrated that immediate solubility storage of CO_2 is possible using the novel water injection technique developed at the site. CO_2 storage in a stable mineral phase within basalt formations was also demonstrated and on much shorter timescales (months to a year) than expected. The implications from this study are far reaching given the vast distribution of basalt deposits globally. The solubility trapping prevents the formation of a buoyant CO_2 in a reservoir reducing the risk of leakage. Once the CO_2 has been precipitated in a mineral phase, monitoring requirements should be less onerous as there is limited leakage risk. This increases safety and potentially reduces costs.

The volumes of water required may limit potential large-scale basalt storage onshore as groundwater resources are limited in many areas. Research currently implies that seawater could be used, which would be readily available offshore but this option has not been tested.

The Wallula site injected pure supercritical CO_2 and although instant dissolution trapping was not implemented, mineral trapping still occurred within two years, much faster than the 5 to 10 year predictions made via models and simulations.

The global distribution of flood basalts offer very large CO_2 storage potential provided the technical challenges can be addressed with further pilot and demonstration scale evaluation.

Although many flood basalt provinces are extensive they are often remote from large point sources of CO₂. This study has included two examples of large (2,000 MW) coal-fired power stations in comparatively close proximity to basalt formations.

8. Recommendations

- To further the use of basalt formations for CCS storage, potential storage volumes need to be quantified in detail. These capacity estimations could be undertaken in countries such as India and South Africa where there is the biggest potential benefit. A study matching CO₂ sources to potential basaltic storage basins would also be beneficial.
- Further cost analysis is recommended given the difference in CO₂ purity at CarbFix and the reduced monitoring scheme. If a significant cost reduction is possible this needs to be demonstrated at larger scale.
- Future reviews will also be required when more detailed analysis is published from the Wallula Project site.
- Studies into the use of seawater for injection with CO₂ are also required as many storage sites may not be able to depend on locally sourced groundwater. An assessment of the chemical reactions that may be altered and the impact on the rate of reactions needs to be analysed.



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IEA Greenhouse Gas R&D Programme

Pure Offices, Cheltenham Office Park, Hatherley Lane, Cheltenham, Glos. GL51 6SH, UK

Tel: +44 1242 802911 mail@ieaghg.org www.ieaghg.org