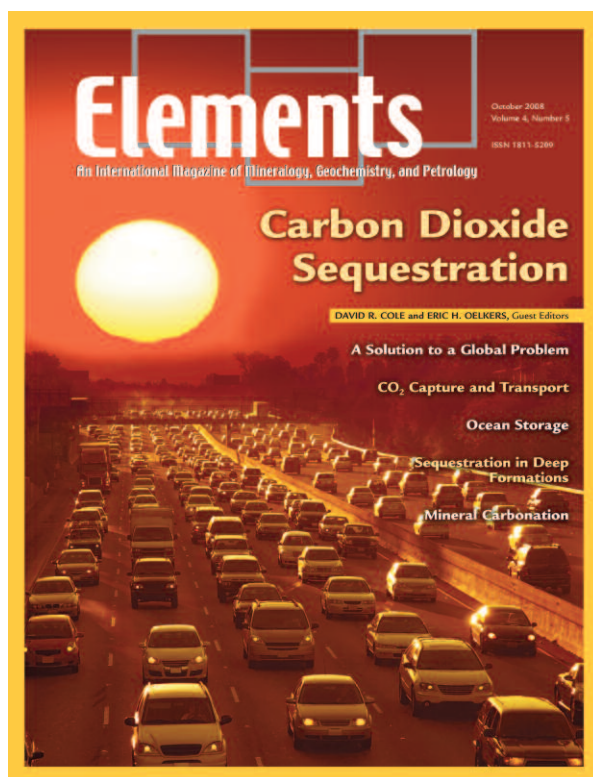


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Mineral Carbonation of CO₂



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A survey of the global carbon reservoirs suggests that the most stable, long-term storage mechanism for atmospheric CO₂ is the formation of carbonate minerals such as calcite, dolomite and magnesite. The feasibility is demonstrated by the proportion of terrestrial carbon bound in these minerals: at least 40,000 times more carbon is present in carbonate rocks than in the atmosphere. Atmospheric carbon can be transformed into carbonate minerals either ex situ, as part of an industrial process, or in situ, by injection into geological formations where the elements required for carbonate-mineral formation are present. Many challenges in mineral carbonation remain to be resolved. They include overcoming the slow kinetics of mineral–fluid reactions, dealing with the large volume of source material required and reducing the energy needed to hasten the carbonation process. To address these challenges, several pilot studies have been launched, including the CarbFix program in Iceland. The aim of CarbFix is to inject CO₂ into permeable basaltic rocks in an attempt to form carbonate minerals directly through a coupled dissolution–precipitation process.

KEYWORDS: CO₂ sequestration, mineral carbonation, mineralogic storage, basalt carbonation

INTRODUCTION

Mineral carbonation is the fixation of CO₂ as stable carbonate minerals, such as calcite (CaCO₃), dolomite (Ca_{0.5}Mg_{0.5}CO₃), magnesite (MgCO₃) and siderite (FeCO₃) (Metz et al. 2005). Insight into the potential effectiveness of storing atmospheric CO₂ as carbonate minerals is demonstrated by the relative volume of these reservoirs worldwide. The total mass of carbon currently in the atmosphere is approximately 800 gigatons (Gt). In contrast, ~39,000,000 Gt of carbon are currently present in carbonate rocks, such as marble, limestone and chalk, in the Earth's crust (cf. Oelkers and Cole 2008 this issue). In nature, the formation of carbonate minerals from atmospheric CO₂ is one of the major processes in the long-term global carbon cycle (Berner et al. 1983). Gaillardet et al. (1999) stated that about 0.1 Gt of carbon per year is bound by silicate-mineral weathering throughout the world; at this rate, the global atmospheric CO₂ inventory would be consumed after about 8000 years.

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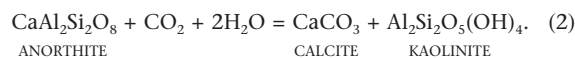
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Mineral carbonation requires combining CO₂ with metals to form carbonate minerals. With few exceptions, the required metals are divalent cations, including Ca²⁺, Mg²⁺ and Fe²⁺. One of the major challenges in the mineral sequestration of CO₂ is, therefore, to obtain these cations. The most abundant cation source is silicate minerals. Carbonate phases are energetically favoured to form from the interaction of CO₂ with such silicate phases as forsterite and anorthite (Seifritz 1990; Lackner et al. 1995; Lackner 2002) in accord with reactions such as:



and



Such reactions have been observed in nature and in the laboratory (e.g. Giammar et al. 2005; McGrail et al. 2006). An alternative to the formation of divalent-cation carbonate minerals, which could eliminate the risk of forming pore-clogging Al-bearing clay minerals, is dawsonite [NaAl(CO₃)(OH)₂], but this compound has high reactivity and relatively low stability, so it is not a good candidate for long-term CO₂ storage (Hellevang et al. 2005; Bénézech et al. 2007).

EX SITU MINERAL CARBONATION

Although the formation of stable carbonate phases by reacting CO₂ with silicate minerals is thermodynamically favourable, it is not straightforward on an industrial scale. There are three major challenges: (1) the mammoth scale of an ex situ CO₂ mineralization effort, (2) the need to accelerate the rate of carbonate formation to make it efficient and (3) the large energy costs (cf. Rubin 2008 this issue).

Scale

According to the U.S. Environmental Protection Agency (EPA) E-Grid, an average coal-fired power plant produces 4 million MWh of electricity per year (DOE/EPA 2000). Coal power plants in the US produce approximately one metric

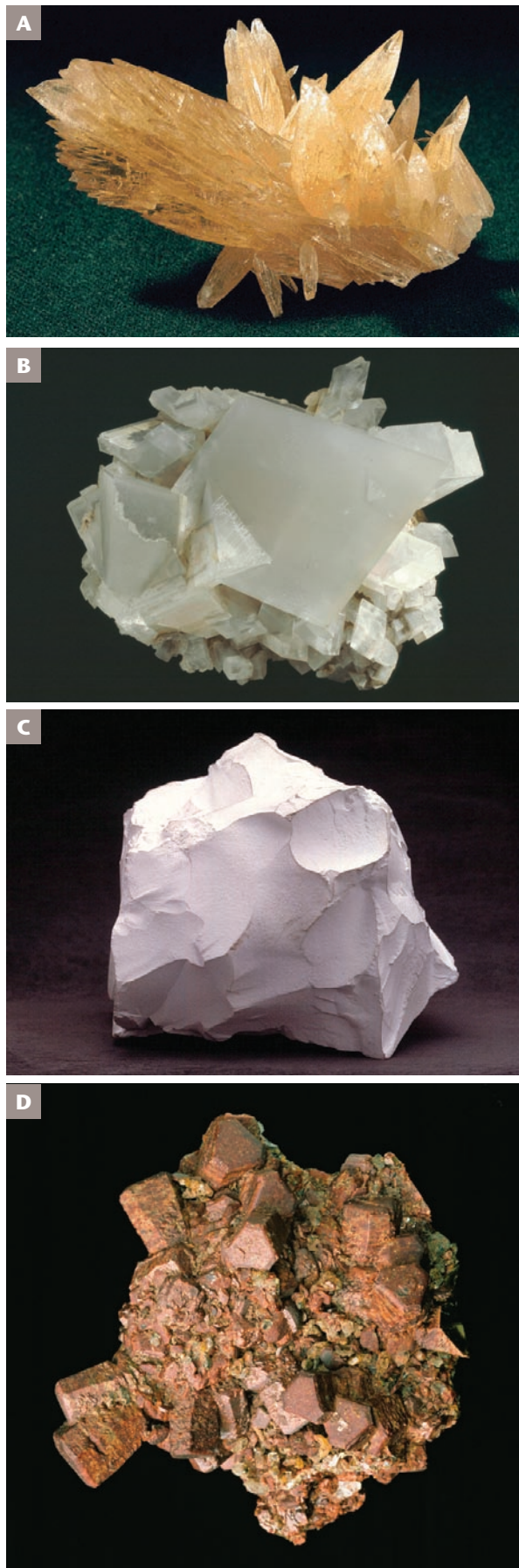


FIGURE 1 Potential mineral hosts of CO₂: (A) calcite, (B) dolomite, (C) magnesite, (D) siderite. COPYRIGHT NHM PICTURE LIBRARY

ton of CO₂ per MWh (DOE/EPA 2000). Based on this emission factor, a 4-million-MWh coal-fired plant emits 4 million tons of CO₂ or 1.1 Mt (1.1×10^3 Gt) of carbon per year.

TABLE 1 lists potential sources of divalent metals required for carbonation; FIGURE 1 and TABLE 2 show the potential CO₂ mineral hosts. The most efficient mineral for sequestering carbon is forsterite, but 6.4 Mt of forsterite would be required to fix the 4 Mt of CO₂ emitted annually from a typical coal-fired power plant. This process would create 2.6 million cubic metres of magnesite, equal to the volume of a cube 140 metres on a side. Note, however, that natural rocks do not contain only the minerals listed in TABLE 1. Numerous other minerals may be present, including those that are poor in divalent cations and those that provoke precipitation of other minerals, such as clays, that compete for the available dissolved metals. As a result, the total mass of rock required to fix CO₂ is usually substantially larger than that listed in this table.

Accelerating Carbonation Rates

In nature, mineral carbonation reactions such as equations 1 and 2 are thermodynamically favoured but proceed slowly. Both in nature and in most mineral carbonation scenarios, the metal-silicate mineral dissolves, and then, through a coupled reaction, a metal carbonate precipitates. In most cases, silicate-phase dissolution rates are slower than the corresponding carbonate precipitation rates, so hastening dissolution is the key to optimization. The dissolution rates of forsterite, wollastonite, serpentine, anorthite and basaltic glass are accelerated by increasing the water-mineral interface area, by increasing temperature and by adding acid or, in some cases, base (cf. Oelkers 2001; Carroll and Knauss 2005). Organic acids have also been observed to accelerate silicate dissolution rates (Golubev and Pokrovsky 2006). A number of studies have attempted to optimize ex situ carbonation methods by increasing temperature, by grinding the silicate source materials and by dissolving them in acidic solutions (e.g. Huijgen et al. 2006).

Energy Penalty and Costs

Ex situ formation of carbonate minerals requires energy and money to transport suitable initial material to a 'carbonation reactor', to grind this material, to heat the reactor system and to store or dispose of the solid CO₂-rich end product. The overall energy costs (or penalty; cf. Rubin 2008) depend on many factors, including the transport distance, the cation content and solubility of the original silicate rock, the degree of grinding required and the quality of the CO₂ stream to be treated. One estimate was reported by Gerdemann et al. (2007), who studied the reaction of Ca-, Fe- and Mg-silicate minerals. Their evaluation of resources, kinetics, process development and costs for each step in the

TABLE 1 SOME POTENTIAL SOURCE MINERALS FOR CARBON MINERALIZATION

SOLID	CHEMICAL FORMULA	Tons required to sequester 1 ton of carbon
Wollastonite	CaSiO ₃	9.68 ^a
Forsterite	Mg ₂ SiO ₄	5.86 ^b
Serpentine/chrysotile	Mg ₃ Si ₂ O ₅ (OH) ₄	7.69 ^b
Anorthite	CaAl ₂ Si ₂ O ₈	23.1 ^a
Basaltic glass	Na _{0.08} K _{0.008} Fe(II) _{0.17} Mg _{0.28} Ca _{0.26} Al _{0.36} Fe(III) _{0.02} SiTi _{0.02} O _{3.45}	8.76 ^c

^a as calcite; ^b as magnesite; ^c assuming all Ca, Mg and Fe are converted into calcite, magnesite and siderite

process suggested a total price of ~\$54 per ton of CO₂ sequestered. However, they also concluded that the scale of ex situ mineral carbonation operations needed to immobilize the CO₂ emissions from a 1 GW coal-fired power plant would require moving 55,000 tons of rock per year; the mass of rock that would have to be mined, transported and eventually stored could make such operations impractical.

Another process proposed for ex situ mineral carbonation is to combine CO₂ with fly ash, which is the solid residue generated from coal combustion when lime is used to neutralize the gas stream in the chimney. Further neutralization of the ash with CO₂ produces carbonate minerals (Soong et al. 2005). Fly ash may contain as much as 30% CaO and Fe₂O₃, depending on the impurities in the coal itself and its burning efficiency, making it an effective source of metal cations. However, limited supplies of fly ash make this option unfeasible at a large scale. US coal-fired power plants produced 71.1 Mt of fly ash in 2005, which is less than 5 mass% of the CO₂ emitted annually in the US. Note also that the production of lime, which is the source of much of the CaO present in the fly ash, releases substantial CO₂ to the atmosphere. Nevertheless, although it cannot provide a complete CO₂-sequestration solution, removal by fly ash could aid CO₂ immobilization efforts while removing particulate material and acid gases (CO₂, NO_x, SO_x) from the chimney gas.

IN SITU MINERAL CARBONATION

In situ mineral carbonation overcomes some of the limitations of an ex situ process because in this approach CO₂ is injected directly into porous rocks in the subsurface, where it can react directly with the host rock. In situ carbonation eliminates the need for transport of reactants in and end products out, and may provide heat to accelerate the carbonation process. The choice of host rock, however, is critical. It must contain easily dissolved metal cations and have sufficient permeability and pore volume to store the injected CO₂ and carbonate-mineral products.

A number of reactive-transport calculations has been made to assess the degree to which in situ CO₂ mineralization is possible within sandstones and sandstone-shale sequences (e.g. Xu et al. 2005). Such calculations suggest that both dawsonite and iron-bearing carbonates could precipitate, depending on the composition of the host rock and, in particular, the presence of iron(II)-bearing minerals such as chlorite. Such reactions, however, appear to be relatively slow in response to sluggish silicate dissolution rates; mineral carbonation in such rocks could require 100,000 years or more to produce 90 kg of carbonate minerals per cubic metre of sandstone, if sufficient Al, Na and Fe are present in the rock.

In contrast, in situ mineralization may be far more effective in basalt or ultramafic rock (e.g. McGrail et al. 2006; Matter et al. 2007), which are rich in divalent cations and poor in silica. A typical basalt contains 7–10 wt% Ca, 5–6 wt% Mg and 7–10 wt% Fe, and these metals are readily liberated by reaction with CO₂-rich water.

There is diverse evidence demonstrating the potential of CO₂ sequestration in mafic and ultramafic rocks. First, despite the fact that less than 10% of the Earth's continental surface is covered by basalt, it takes up ~33% of all the CO₂ consumed during natural weathering of Earth's surface silicates (Dessert et al. 2003). Second, the flux of Mg and Ca liberated when CO₂-rich water reacts with basalt is as much as two orders of magnitude larger than when they react with other, more crystalline or silica-rich rocks (Wolff-Boenisch et al. 2006). Third, several well-documented examples of carbonation

of basaltic rocks are known, such as through hydrothermal alteration (Gudmundsson and Arnórsson 2002) and surface weathering (Gislason and Eugster 1987; Gislason et al. 1996; Gislason et al. 2006). Fourth, enormous volumes of mafic and ultramafic rocks are present on the Earth's surface. For example, the Columbia River basalts have a volume in excess of 200,000 km³ and the Siberian basalts, 1,000,000 km³. The locations of some of the major continental basalts are shown in FIGURE 2. These large volumes have a correspondingly large CO₂-sequestration capacity: McGrail et al. (2006) estimated that the Columbia River basalts alone have the capacity to sequester in excess of 100 Gt of CO₂. Many lava flows are porous and permeable and have enormous storage capacity. Basalts also dominate the seafloor, providing vast in situ carbon-mineralization sites throughout the world (Goldberg et al. 2008). Thus, storage in basalts is now considered to be among the best of the options for sequestering CO₂ (O'Connor et al. 2003).

Further insight into in situ mineral carbonation in basalts and ultramafic rocks comes from the results of reactive-transport modeling. Marini (2007) simulated the interaction of injected CO₂ with a number of potential host rocks. Results for CO₂ in a basaltic-glass reservoir are shown in FIGURE 3. Dissolution of basaltic glass by carbonated water promotes the formation of chalcedony (SiO₂), goethite (FeOOH) and the clay mineral kaolinite. The model suggests that after ~1 year, siderite precipitates, followed by dolomite, dawsonite and magnesite. Substantial CO₂ mineralization is evident after 10–100 years of reaction. The volume of these secondary phases is, however, substantially greater than that of the basaltic glass that was dissolved to make them, leading to the potential of these phases to clog the available pore spaces and, in some cases, to substantially decrease the permeability of the host rock (Gysi and Stefánsson 2008).

In situ mineral carbonation in basalt or ultramafic rocks is not without its challenges. These include the availability of water for carbonation, the need for impermeable cap rocks, and the possible mobilization of trace and toxic metals.

TABLE 2 CARBONATE PHASES FOR POTENTIAL MINERAL STORAGE OF CO₂

Mineral	Formula	Mass produced per ton C (ton)	Volume produced per ton C (m ³)
Calcite	CaCO ₃	8.34	3.08
Magnesite	MgCO ₃	7.02	2.36
Dawsonite	NaAl(CO ₃)(OH) ₂	12.00	4.95
Siderite	FeCO ₃	9.65	2.49
Ankerite	Ca(Fe,Mg)(CO ₃) ₂	8.60	2.81

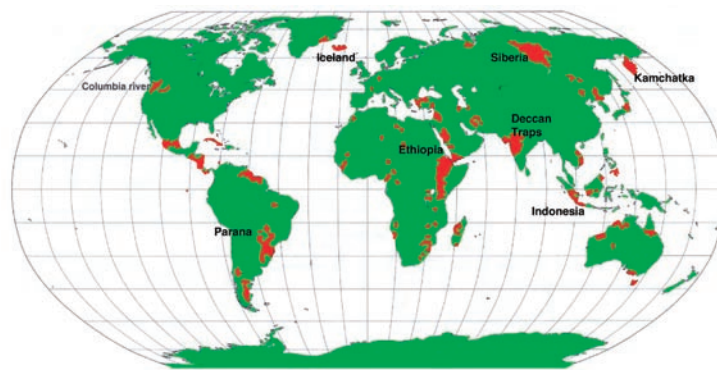


FIGURE 2 Locations of continental basalts that could serve as in situ mineral carbonation sites

AVAILABILITY OF WATER

In situ mineral carbonation requires that the injected CO₂ dissolve into an aqueous solution. CO₂ can be dissolved in water prior to its injection or injected into the target rock formation so that it can dissolve directly into the existing groundwater. Dissolving CO₂ prior to its injection requires large quantities of water. To dissolve one ton of CO₂ at 25 bars partial pressure and 25°C requires ~27 tons of water. This quantity increases to ~341 tons of water at a CO₂ partial pressure of 2 bars. If such large volumes of water are available, CO₂ dissolution can be accomplished at the borehole or during injection. Injecting large quantities of CO₂-rich water requires substantial well-bore permeability at the locality of the bore hole. Injection costs depend on the amount of energy required to pump the CO₂-rich water down the hole. Some of these challenges may be overcome by injecting CO₂ into oceanic basalts. This procedure offers

two advantages: (1) a large volume of water for CO₂ dissolution is readily available (Brady and Gislason 1997) and (2) such basalts may be covered by low-permeability sediments, which serve as a seal to slow water escape, allowing time for reaction (Goldberg et al. 2008).

Direct injection of CO₂ overcomes the difficulty of finding sufficient water, but leads to other problems. If injected as a separate phase, CO₂ must dissolve into the groundwater before carbonation can begin. CO₂ diffusion into groundwater can be a slow process, depending on system hydrology and the chemical composition of the groundwater.

Cap Rock Impermeability

Because both CO₂ dissolution into groundwater and coupled mineral dissolution-precipitation reactions leading to carbon mineralization may be slow, an impermeable cap rock is essential to keep the dissolved CO₂ in contact with the reactive host rock for sufficient time to allow carbonation. Cap rocks are common in oilfield reservoirs but may be missing over a basalt or ultramafic formation. A further complication is that these rock types are commonly fractured. The permeability of the cap rock itself may be altered by its reactions with the injected CO₂, leading to leaks in the originally impermeable barrier (Gaus et al. 2005).

Trace- and Toxic-Metal Mobility

The dissolution of CO₂ into water produces carbonic acid, which reacts with the host rocks to liberate the cations necessary to create carbonate minerals. These reactions may also release trace and toxic metals into solution, allowing them to migrate. The degree to which these metals move with the formation water depends on the hydrology and chemistry of the system. Flaathen and Gislason (2007) reported that trace and toxic metals released from the interaction of basalt and CO₂-charged water are readily immobilized as further reactions increase groundwater pH. The degree to which these metals are immobilized in sandstone, however, remains unclear.

The CarbFix Project

In an attempt to assess the feasibility of in situ CO₂ mineralization in basaltic rocks, a field-scale project, CarbFix, is scheduled to begin during 2009 at Hellisheidi, Iceland (www.carbfix.com). The injection site (FIG. 4) is adjacent to a new geothermal power plant, which will supply up to 30,000 tons of CO₂ per year for injection into subsurface basalts at a depth of 400–700 m and a temperature of ~30°C. The Hellisheidi site was chosen for several reasons, including a local source of CO₂, the availability of several strategically located wells for monitoring the chemical evolution of the groundwater, and the proximity to infrastructure through Reykjavik Energy, a CarbFix partner. CO₂ will be injected at a partial pressure of 25 bars and will be dissolved in water during injection. It is anticipated that the results of this project will be used to optimize the in situ carbon mineralization process, enabling in situ carbonation at sites throughout the world.

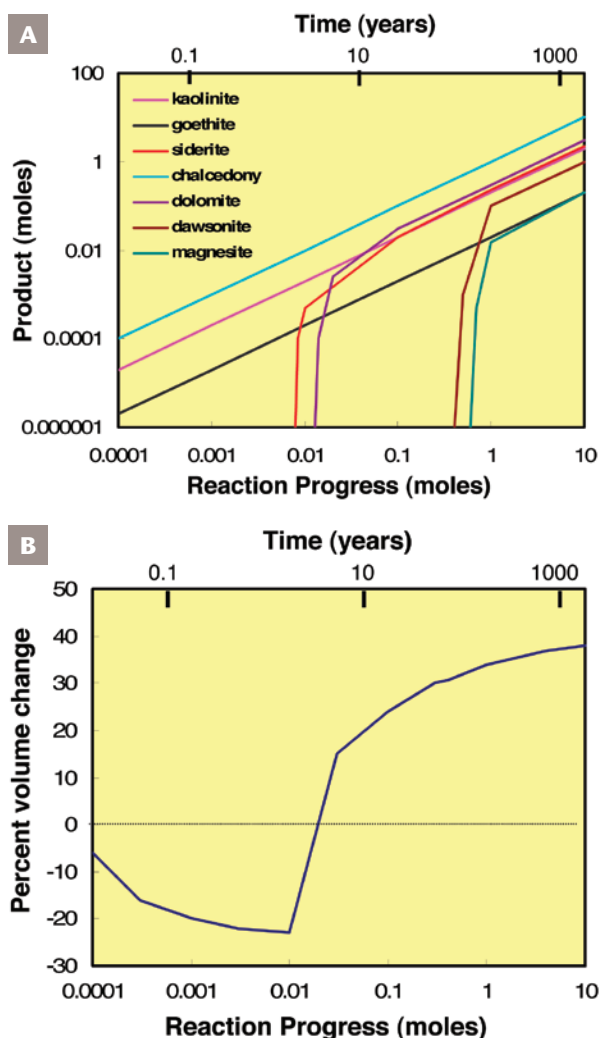


FIGURE 3 Results of reactive-transport calculations for the injection of CO₂ at a pressure of 100 bars into a basaltic-glass reservoir at 60°C (after Marini 2007): (A) mass of secondary minerals formed and (B) change in rock volume as a function of reaction progress and time. Reaction progress is presented as the number of moles of basaltic glass dissolved. For example, after 10 years of the interaction of the CO₂-rich fluid with a basaltic-glass reservoir, one might expect that 0.06 moles of glass had dissolved, leading to the formation of 0.05 moles of chalcedony, 5×10^{-3} moles of kaolinite, 5×10^{-3} moles of dolomite, 4×10^{-3} moles of siderite and 5×10^{-4} moles of goethite. These reactions would increase net volume by 20%, potentially clogging existing pores. Note that these are the results of model calculations, which provide useful predictions, but different minerals could precipitate in the natural systems.

CONCLUSIONS AND PERSPECTIVES

In situ CO₂ mineralization offers the potential for long-term, safe CO₂ storage at a reasonable price. Optimizing this technology, however, will require a multifaceted effort, including field-scale pilot studies, to better characterize the rates of mineral carbonation reactions and the fate and consequences of injecting CO₂ into reactive silicate rocks. Public acceptance and government/industry support will require development of new monitoring techniques to demonstrate that the injected CO₂ has indeed been fixed

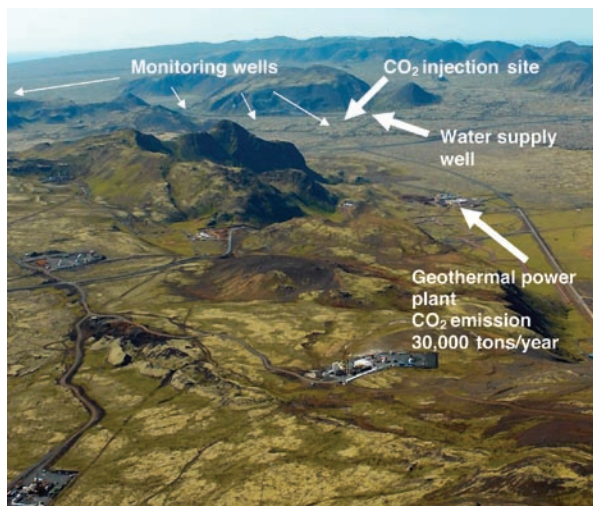


FIGURE 4 The field-scale, in situ basalt-carbonation pilot plant in Hellisheiði, Iceland. PHOTO BY MATS WIBERLUND

in carbonate rocks, with minimal risk that it will leak to the atmosphere. Such monitoring techniques may involve developing chemical or isotopic fingerprinting methods that can characterize subsurface carbonation. They will also require improved techniques to quantitatively define subsurface fluid flow paths and, eventually, sites of blockage. Thus, a large effort by the scientific community is still required to ensure that this sequestration method is economically and technologically viable.

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