Modelling a basalt reactor for direct air CO₂ capture

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Received: 21 September 2021 / Accepted: 6 March 2022 / Published online: 19 March 2022

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Abstract
Ground basalt has been used as mineral fertilizer since the early thirties. Ground basalt captures CO₂ from the atmosphere and the soil pore space, raises the soil pH and reduces ocean acidification. One tonne of basalt captures 0.153–0.165 tonne CO₂, depending on infiltration rate (400–1200 mm/a), reactive surface area (3.7–15 m²/g) and CO₂ partial pressure (41.1–3000 Pa). When the infiltration rate is high (1200 mm/a), the CO₂ capture capacity of basalt is exhausted after 9.5–11.4 years. When the infiltration rate is low (400 mm/a), the capture capacity is exhausted after 28.2–33.1 years. With the exhaustion of the capture capacity, the newly formed carbonates that sequestered CO₂ start dissolving. The dissolution is complete after 34.9–101.7 years, depending on infiltration rate, reactive surface area and CO₂ partial pressure. The reaction products are transported to the ocean via surface waters. The degree to which the fugacity of CO₂ controlled by the Henry constant exceeds the fugacity of atmospheric CO₂ along the travel path depends on many unknowns. Thus, it is impossible to reliably predict to which degree the captured CO₂ is recycled to the atmosphere, if it is recycled at all.

Keywords Climate change · Carbon dioxide · CO₂ · Basalt fertilizer · Ocean acidification · Numerical model

Introduction
Ground basalt added to agricultural fertilizer captures atmospheric carbon dioxide (CO₂), raises the soil pH, reduces ocean acidification and supplies important nutrients such as magnesium, potassium, calcium, iron and phosphorus. Ground silicate rock has been used as mineral fertilizer since the early thirties (Hilf 1938; de Villiers 1961; Gillman et al. 2002; van Straaten 2006; Anda et al. 2009). More recently, CO₂ sequestration by silicate rock has been investigated by incubation and infiltration experiments, which had reaction periods in the range of 3–12 months (ten Berge et al. 2012; Dietzen et al. 2018; Kelland et al. 2020; Amann et al. 2020). These experiments exploited only a minute fraction of the CO₂ sequestration potential. However, reactive-transport modelling with the TOUGHREACT software is capable of covering the full lifetime of a basalt reactor. The design of models with various environmental parameters is the subject of this paper.

The weathering of silicate rock involves the reaction with dissolved carbon species, the activities of which are controlled by the thermodynamic equilibrium between CO₂ gas in the atmosphere and soil, on one hand, and liquid CO₂, on the other hand. This is a natural process, which controls the CO₂ level of the atmosphere on the geological time scale. The only way to enhance this natural process is to increase the reactive surface area of the silicate rock by grinding.

In terms of CO₂ sequestration potential, basalt is unique. Basalt volcanic and subvolcanic rock occupies 5.6 × 10⁶ km² or 3.5% of the land surface of the Earth (Fig. 1; Hartmann and Moosdorf 2012). Important occurrences are located in South Africa, Russia, Brazil, Ethiopia, India, the USA, Iceland, Australia, and some neighbouring countries.

The capture of CO₂ from the atmosphere via chemical reaction with ground basalt rock added to agricultural fertilizer implies additional costs. These are investment costs rather than operational costs because of the long lifetime of a basalt reactor. Basalt amendment typically would constitute less than one third of the price of high-yielding agricultural land under favourable transport conditions (transport by lorry, train and/or ship). Combining the benefits of high yield induced by basalt amendment and income from CO₂ emission certificates may lead to an economically viable situation.
Basalt amendment shares the high environmental costs of climate neutrality with other techniques such as storage of electrical energy or hydrogen production. These environmental costs arise from the mining of immense amount of rock and minerals needed for the setup-up of the emission-reducing installations.

The purpose of this research is to predict the performance of the basalt reactor as a function of reactive surface area, CO₂ partial pressure and rainwater infiltration. The research is meant to guide decision-makers in a more reliable way than previous studies with a more general approach (Taylor et al. 2015; Taylor et al. 2017; Strefler et al. 2018; Lefebvre et al. 2019; Beerling et al. 2020a).

Materials and methods

TOUGHREACT computer code

All models have been calculated with TOUGHREACT version 3. This a numerical simulation programme for chemically reactive flows of multi-phase fluids in porous and fractured media (Xu et al. 2014a). Interactions between mineral assemblages and fluids can occur under local equilibrium or via kinetic rates. The reaction rate is a function of the mineral saturation ratio and is calculated with the rate expression of Lasaga et al. (1994). This is a chemical inhibition term, which slows the dissolution/precipitation rate as the chemical state of the fluid approaches chemical equilibrium with respect to the dissolving/precipitating mineral phase.

Flow model

Base case

A structured orthogonal model mesh with three elements and horizontal dimensions of 1 × 1 m is used (Table 1). The top and bottom elements have a volume of $10^{5.2}$ m³. Thus, Dirichlet conditions are imposed, i.e., the thermodynamic conditions of the top and bottom element do not change at all. The intermediate element has a volume that represents

![Fig. 1 Occurrences of basaltic volcanic and subvolcanic rock (Hartmann and Moodorf 2012). Equal-area Mollweide projection](image)

| Table 1 Flow model setup |
|--------------------------|
| Simulation period (a)    | 101.7 |
| Model length/width (m)   | 1/1   |
| Temperature (°C)         | 25    |
| Total pressure (Pa)      | 0.999947×10⁵–1×10⁶ |
| CO₂ partial pressure (Pa) | 41.1–3000 |
| Porosity (–)             | 0.3   |
| Permeability (m²)        | $10^{-13}$ |
| Basalt grain density (kg/m³) | 2800 |
| Basalt reactive surface area (m²/g) | 3.7–15 |
| Basalt application rate (t/ha) | 100  |
| Infiltration rate (mm/a) | 400–1200 |
100 tonnes of ground basalt distributed over an area of 1 hectare (0.005 m³). The total pressure at the top element is 10⁵ Pa, and the partial pressure of CO₂ gas is set at 41.1 Pa, which is actual atmospheric partial pressure at standard conditions (10⁵ Pa, 25°C). The bottom element has a total pressure of 0.999982 × 10⁵, 0.999965 × 10⁵ or 0.999947 × 10⁵ Pa, which serves to maintain an infiltration rate of 400, 800 or 1200 mm/a, respectively. These infiltration rates characterize mid-latitude, subtropical and tropical climate conditions.

The rock grain density has the typical value of Columbia River basalt, U.S.A. (2.8 g/cm³; DOE 1982). The remaining material parameters are taken from a CO₂ sequestration test case of Xu et al. (2014b).

**Sensitivity cases**

The sensitivity cases have the same setup as that of the base case but use soil CO₂ partial pressure instead of atmospheric CO₂ partial pressure. The lower limit (3 × 10² Pa) and upper limit (3 × 10³ Pa) of the range given by Bohn et al. (2001) are applied.

**Reactive transport model**

**Base case**

The transport conditions are simulated for ground rock with a reactive surface area of 7.4 m²/g determined for a grind size P₈₀ = 1250 μm (80% sieve passing size) for crushed basalt using the N₂ adsorption method (Kelland et al. 2020) and calculated with the BET equation named after Brunauer, Emmett, and Teller who developed the theory. The initial and boundary water composition is that of rainwater collected in the tropical Pune area, India (Momin et al. 2005), supplemented with Fe and Si analyses of rainwater collected in the Panipat area, India (Bharti et al. 2017), and an Al analysis of rainwater collected in the Bandung area, Indonesia (Hasan et al. 2017) (Table 2). The temperature is 25 °C. The initial and boundary partial pressure of CO₂ gas is set at 41.1 Pa, which is actual atmospheric partial pressure at standard conditions (10⁵ Pa, 25°C). This setup defines a total aqueous carbon concentration of 1.3 × 10⁻⁵ mol C per kg H₂O (molal).

The glass fraction of the basalt is 0.25, which is a typical value for Columbia River basalt, USA, such as that used by Kelland et al. (2020). The proportion of the crystallised phases corresponds to the mineralogical composition of the basalt used by Pollyea and Rimstidt (2017) (Table 3). A volume fraction of 0.02, consisting of TiO₂ and P₂O₅ components, is neglected, i.e., assumed to be non-reactive. A simplified set of reaction products is used (Pollyea and Rimstidt 2017): calcite, siderite, magnesite (i.e. carbonates representative of ankerite-dolomite solid solution; Reeder and Dollase 1989), amorphous SiO₂, Ca-montmorillonite, Na-montmorillonite and illite (i.e. Al-silicates representative of mixed-layer minerals solid solution; Meunier and Velde 1989). There are no kinetic data for solid solutions; therefore, the available data for minerals with a fixed composition must be used. Basalt glass (Pollyea and Rimstidt 2017) can only dissolve. Siderite (Knauss et al. 2005) and the remaining components (Palandri and Kharaka 2004) dissolve and precipitate under kinetic constraints. Illite is assumed to have the same rate constants as montmorillonite. The precipitation rates equal dissolution rates. The initial volume fraction of all reaction products is zero.

The calculations are performed with the THERMODDEM thermodynamic database (Blanc et al. 2007), supplemented by data for basalt glass (Pollyea and Rimstidt 2017). All aqueous species of the elements shown in Table 2 were allowed to take part in the reactions.

**Sensitivity cases**

The sensitivity cases have the same setup as that of the base case but use the reactive surface area of the base case multiplied and divided by 2. The corresponding values are 15 and 3.7 m²/g, respectively.

**Results**

**Base case**

The performance of the basalt reactor is monitored by the efficiency ratio. This is the CO₂/basalt weight ratio corresponding to the fraction of basalt that can be converted into carbonate minerals. The theoretical efficiency ratio is 0.211 but this value is not achievable under realistic assumptions, because basalt can react with water without the participation of aqueous carbon species.

Figure 2 shows that the efficiency ratio varies little with infiltration (0.161–0.165) at the end of the sequestration

| pH (–) | 5.7 |
|-------|-----|
| Al (mol/kgH₂O) | 9.7 × 10⁻¹⁰ |
| C (mol/kgH₂O) | 1.3 × 10⁻⁵ |
| Ca (mol/kgH₂O) | 1.2 × 10⁻⁴ |
| Cl (mol/kgH₂O) | 8.9 × 10⁻⁵ |
| Fe (mol/kgH₂O) | 2.3 × 10⁻⁹ |
| K (mol/kgH₂O) | 1.3 × 10⁻⁵ |
| Mg (mol/kgH₂O) | 2.4 × 10⁻⁵ |
| Na (mol/kgH₂O) | 4.4 × 10⁻⁵ |
| O₂ (mol/kgH₂O) | 2.7 × 10⁻⁴ |
| Si (mol/kgH₂O) | 1.2 × 10⁻⁷ |
But the time span for reaching the end of the sequestration period is strongly influenced by the infiltration rate. At tropical conditions (1200 mm/a), the end point is reached after 11.2 years. At mid-latitude conditions (400 mm/a), the end point is reached after 33.1 years.

The end point is reached when all basalt glass and feldspar have dissolved (Fig. 3). Although dissolution of plagioclase and pyroxene continue beyond the end point, the activities of aqueous Ca, Mg and Fe species remain too low for stabilizing previously precipitated carbonate minerals. The net effect is the reduction of the SmCO2 value (mass of CO2 sequestered in solid mineral phases). All previously precipitated carbonate minerals are completely dissolved after 36.5 years and 101.7 years in the 1200 mm/a and 400 mm/a scenarios, respectively.

It is implicitly assumed that CO2 originally sequestered via solid carbonate phases is not recycled to the atmosphere but stored in the geosphere and hydrosphere. Nevertheless, field studies that support such an assumption have not been conducted yet (Kelland et al. 2020).

**Sensitivity cases**

Multiplying the reactive surface area of the base case (7.4 m²/g) by 2 has hardly any positive effect on the efficiency ratio (Fig. 2). In contrast, dividing the base case value by 2 has a significant negative effect on the efficiency ratio especially when the infiltration rate is high. This implies that there is no point in finer grinding to achieve a reactive surface area > 7.4 m²/g. However, coarser grinding resulting in a reactive surface area < 7.4 m²/g risks to jeopardize the performance of the basalt reactor especially under tropical conditions with a high infiltration rate.

The variations of the CO2 partial pressure have little influence on the efficiency ratio at the end of the sequestration period (Fig. 4). But the time span for reaching the end of

| Mineral/glass | Chemical composition | Initial volume fraction (m³/m³) | Reactive surface area (m²/g) |
|---------------|----------------------|-------------------------------|-----------------------------|
| Amorphous SiO₂ | SiO₂                 | 0.0                           | 3.7–15                      |
| Illite        | K₀.₈₅Si₃.₁₅ Al₂.₅₅O₆(OH)₂ | 0.0                           | 3.7–15                      |
| Calcite       | CaCO₃                | 0.0                           | 3.7–15                      |
| Siderite      | FeCO₃                | 0.0                           | 3.7–15                      |
| Magnesite     | MgCO₃                | 0.0                           | 3.7–15                      |
| Ca-montmorillonite | Ca₉.₁₇Al₁.₆₈Mg₀.₃₅Si₃.₉₉O₁₅(OH)₂ | 0.0                           | 3.7–15                      |
| Na-montmorillonite | Na₀.₃₃Al₁.₉₆Mg₀.₃₅Si₃.₉₉O₁₅(OH)₂ | 0.0                           | 3.7–15                      |
| Goethite      | Fe₂(OH)             | 0.0                           | 3.7–15                      |
| Glass         | SiAl₃.₃₅Fe(II)₀.₁₈₉Mg₀.₃₅Ca₀.₂₆₆Na₀.₀₇₉K₀.₀₈₆O₁₅ | 0.25                          | 3.7–15                      |
| Pyroxene      | CaAl₂O₄             | 0.27                          | 3.7–15                      |
| Plagioclase   | Ca₀.₃₃Na₀.₆₄Al₁.₆₈Si₂.₄O₈ | 0.33                          | 3.7–15                      |
| Forsterite    | Mg₂SiO₄             | 0.13                          | 3.7–15                      |

**Table 3** Reactive transport model setup: Initial volume fractions, reactive surface areas and kinetic properties

| Mineral/glass | Dissolution rate parameters |
|---------------|-----------------------------|
| Acetic mechanism | Neutral mechanism | Base mechanism |
| k₂₅ (mol/m²/s) | Eₐ (KJ/mol) | n(H⁺) | k₂₅ (mol/m²/s) | Eₐ (KJ/mol) | k₂₅ (mol/m²/s) | Eₐ (KJ/mole) | n(H⁺) |
| Amorphous SiO₂ | – | – | – | 5.89 × 10⁻¹³ | 74.5 | – | – | – |
| Illite | 1.95 × 10⁻¹³ | 48.0 | 0.22 | 3.89 × 10⁻¹⁵ | 48.0 | 3.89 × 10⁻¹⁵ | 48.0 | 0.13 |
| Calcite | 0.501 | 14.4 | 1.00 | 1.55 × 10⁻⁶ | 23.5 | 3.31 × 10⁻⁴ | 35.4 | 1.00 |
| Siderite | 9.77 × 10⁻⁴ | 20.9 | 0.90 | 1.26 × 10⁻⁹ | 62.8 | – | – | – |
| Magnesite | 4.17 × 10⁻⁷ | 14.4 | 1.00 | 4.57 × 10⁻¹⁰ | 23.5 | 6.02 × 10⁻⁶ | 62.8 | 1.00 |
| Montmorillonite | 1.95 × 10⁻¹³ | 48.0 | 0.22 | 3.89 × 10⁻¹⁵ | 48.0 | 3.89 × 10⁻¹⁵ | 48.0 | – | – |
| Goethite | – | – | – | 1.15 × 10⁻⁸ | 86.5 | – | – | – |
| Glass | 5.37 × 10⁻⁴ | 39.7 | 1.01 | – | – | 1.00 × 10⁻¹¹ | 38.4 | 0.26 |
| Pyroxene | 1.50 × 10⁻⁷ | 78.0 | 0.70 | 1.07 × 10⁻¹² | 78.0 | – | – | – |
| Plagioclase | 1.35 × 10⁻⁸ | 42.1 | 0.63 | 1.23 × 10⁻¹¹ | 45.2 | – | – | – |
| Forsterite | 1.41 × 10⁻⁷ | 67.2 | 0.47 | 2.29 × 10⁻¹¹ | 79.0 | – | – | – |

⁴k₂₅ is the kinetic rate constant at 25 °C; Eₐ is the Arrhenius activation energy; n(H⁺) is the reaction order with respect to H⁺
CO₂ sequestration decreases significantly when the CO₂ partial pressure is high. For example, the infiltration case with 400 mm/a has the end point at 28.2 years at 3000 Pa partial pressure; the corresponding value for a low partial pressure (41.1 Pa) is 33.1 years. The infiltration case with 1200 mm/a has the end point at 9.5 years; the corresponding value for a low partial pressure (41.1 Pa) is 11.4 years.

Comparison with experiments

The comparison of the TOUGHREACT model with experimental results is not straightforward because these are recorded in terms of release of Mg (± Ca) from dissolving ground rock but not in terms of precipitated solid carbonates. The duration, infiltration rate, type of media and ground rock loading of the experiments are

1. 0.33-year infiltration (766 mm/a) into ground basalt mixed with soil containing cereal plants, the rock loading being 100 t/ha (Kelland et al. 2020)
2. 0.61-year infiltration (213 mm/a) into forsterite-dominated olivine (the most reactive mineral component of basalt) mixed with soil containing ryegrass plants, the rock loading being 204 t/ha (ten Berge et al. 2012)
3. 1-year infiltration (800 mm/a) into ground dunite (an ultrabasic plutonic rock mainly composed of olivine)
mixed with soil containing cereal plants, the rock load-
ing being 220 t/ha (Amann et al. 2020).

The efficiency ratio of the experiments is higher or lower
than that of the TOUGHREACT model (Figs. 2 and 4,
Table 4):

1. The modelled efficiency ratio after 0.33 year infiltration
   is in the range of 0.0018–0.0057, i.e., lower than the
   experimental value (0.013)
2. The modelled efficiency ratio after 0.61 year infiltration
   is in the range of 0.0031–0.011, the maximum value
   being slightly lower than the experimental value (0.013)
3. The modelled efficiency ratio after 1 year infiltration is
   in the range of 0.0051–0.018, i.e., the whole range is
   higher than the experimental value (0.0002).

In summary, the experiments do neither confirm nor con-
tradict the TOUGHREACT model.

Comparison with previous models

Two reactive-transport models with a simulation period that
extends beyond that of the experiments with a basalt reactor
have been published (1 year, Beerling et al. 2020b; 5 years,
Kelland et al. 2020). Owing to the restriction of the software
used (PHREEQC, Parkhurst and Appelo 2013), all precip-
itating phases react under equilibrium conditions. The result
is a strongly non-linear relationship between efficiency ratio
and time. This is in stark contrast to the near-linear rela-
tionship of the TOUGHREACT model with both dissolving
and precipitating phases reacting under kinetic constraints.
Preference should be given to the TOUGHREACT model,
because the PHREEQC model relies on unphysical assump-
tions. All participating minerals precipitate under kinetic
constraints with reaction rates that are at least as low as the
dissolution rates (Palandri and Kharaka 2004, and references
therein).

Conclusion

The modelling of the basalt reactor succeeded in predict-
ing its efficiency ratio (0.153–0.165 tonne CO₂ per tonne
ground rock) and its lifetime (9.5–33.1 a) as a function of the
reactive surface area, CO₂ partial pressure and infiltration
rate. These results are useful for decision-makers. E.g., the
combined production costs and 300 km transportation costs
for ground basalt are $ 82 US per tonne for a grind size of
50 μm (Strefler et al. 2018). Accordingly, the costs for cap-
turing one tonne of CO₂ are in the range from $ 496 US to
$ 536 US. These costs are considerably less than the costs
of the alternative adsorption/absorption method of direct air
CO₂ capture if the obligatory CO₂ underground injection is
included. For the above-ground adsorption/absorption unit
alone, Viebahn et al. (2019) quote $ 540 US per tonne of
captured CO₂ for both the Climeworks installation, Swit-
zerland, and the Carbon Engineering installation, Canada,
a recipient of the Gates Foundation. Including underground
storage in Iceland basalt, Climeworks charges EURO 960 ($
1100 US) per tonne CO₂ compensation in 2021 (Niemann
2021).

The permanent and safe underground storage of CO₂ is an
expensive enterprise with considerable technical, geological
and environmental problems. For underground CO₂ injec-
tion on the large scale, there is no easy solution in sight. To
date, there is no major on-land operation for injecting CO₂
derived from an industrial source except for the petroleum
industry. However, the petroleum industry is a very special
case because CO₂ is obtained as an unwanted by-product
during gas or petroleum exploitation and is reinjected close
to its original source.

The optimal underground target for injecting CO₂ cap-
tured with a physical (non-reactive) method is basalt where
CO₂ is converted into solid carbonates (Schwartz 2020);
whereas non-reactive target rocks such as sandstone imply the risk that injected CO₂ may leak to a near-surface aquifer and pollute the groundwater (Schwartz 2014, and references therein).

Underground CO₂ injection into basalt and mining of basalt for fertilizer production are likely to compete for space if conducted on a large scale. About 3.5% of the Earth’s surface is occupied by basalt but only a minor fraction has the transport infrastructure that allows a reasonably economic use of these basalt resources. In summary, the CO₂ direct air capture is bound to encounter many obstacles. Nevertheless, modelling direct air capture is a useful comparative tool for demonstrating the advantages of reducing greenhouse gas emissions at the source.

Fig. 4 Basalt reactors with a reactive surface area of 7.4 m² and various combinations of infiltration rate and CO₂ partial pressure. Efficiency ratio (CO₂/basalt weight ratio; see text; black solid line; vertical scale text in black) and SmCO₂ (total CO₂ sequestered in mineral phases in kg/m³ medium; red interrupted line; vertical scale text in red) plotted versus time. Efficiency ratios obtained from infiltration experiments (red point symbols; see text) are shown for comparison.
Table 4  Comparison of experiments and models for direct air CO2 capture

| Experiment/model type | Infiltration rate (mm/a) | Grind size | Reactive surface area (m²/g) | Incubation/infiltration period (a) | Efficiency ratio (–) | References |
|-----------------------|-------------------------|------------|-----------------------------|-----------------------------------|----------------------|------------|
| Incubation            |                         |            |                             |                                    |                      |            |
|                        |                         |            |                             |                                    |                      |            |
| Infiltration           |                         |            |                             |                                    |                      |            |
| 213                   | 174                     | P50 = 50 μm | 7.4                         | 0.61                               | 0.013                | Kelland et al. (2012) |
| Infiltration           |                         |            |                             |                                    |                      |            |
| 800                   | 110                     | P73 = 62 μm | 7.4                         | 0.33                               | 0.013                | Kelland et al. (2020) |
| Infiltration           |                         |            |                             |                                    |                      |            |
| 1200                  | 100                     | P1250 = 1250 μm | 7.4                     | 0.161                              |                      | This paper³ |

CO₂ partial pressure 41.1 Pa

Funding  The authors have not disclosed any funding.

Declarations

Conflict of interest  The author has no competing interests.

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