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## Carbon sequestration potential of altered mafic reservoirs

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### Abstract

Porous basaltic aquifers are currently being considered as a key geologic carbon storage host due to its widespread distribution and high reactivity. The co-injection of CO<sub>2</sub> and groundwater into basaltic reservoirs has the potential to mineralize this gas into solid phases within years, thanks to the release of divalent cations. Many natural basalts, however, contain substantial alteration minerals. Here we explore the potential of basalt alteration minerals to provide the Ca to fix injected CO<sub>2</sub> within calcite and/or aragonite. Preliminary results suggest that altered basaltic rocks can provide this Ca as efficiently as fresh basalts at 25 and 100 °C. Further experimental work is ongoing to confirm these findings at different temperatures and as a function of injected fluid chemistry.

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*Keywords:* Carbon sequestration; altered basalt; calcium release rate; epidote

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### 1. Introduction

Emissions of carbon dioxide (CO<sub>2</sub>) into the atmosphere must be substantially reduced and/or carbon capture and storage efforts must be substantially increased to meet the goals of the Paris agreement, which aims to keep the temperature rise “well below 2 °C”, compared to pre-industrial levels [1]. In-situ mineral carbonation is one of the several mitigation strategies proposed to safely store CO<sub>2</sub> over extended time frames [2, 3]. This process may be promoted by the reaction of CO<sub>2</sub>-charged water with the surrounding host rocks in the subsurface to form geologically stable carbonate minerals [4]. This long-term storage option reduces the likelihood of accidental CO<sub>2</sub> release and the need of post-storage monitoring [5, 6], making mineral carbonation one of the most promising trapping mechanisms [7, 8].

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This mineral storage has been found to be particularly efficient in mafic and ultramafic rocks [9-16], such as basalts and peridotites, where the abundance of divalent cations (e.g.  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{Fe}^{2+}$ ) promotes the precipitation of  $\text{CO}_2$  as carbonates. In particular, the findings from the CarbFix project – a pilot field study injecting  $\text{CO}_2$ -charged fluids within a basaltic reservoir in Iceland – show that the time required to mineralize over 95% of the injected  $\text{CO}_2$  into 30 °C basalts was less than two years [17, 18]. However, most of laboratory and field scale results are based on “ideal” scenarios characterized by relatively young and highly reactive rocks, and do not reflect the variety of potential reservoirs spread across our planet, many of which contain abundant alteration minerals.

To address this issue, the reactivity of common fresh and alteration phases are compared. Here, we report the preliminary results of this effort, focusing on altered basaltic rocks.

## 2. Basaltic reservoirs

Basalts offer a potentially huge reservoir capacity for carbon storage, both on-land [3] and off-shore [19]. A critical step to quantifying how efficiently these rocks can mineralize injected  $\text{CO}_2$  is the rate at which they liberate their divalent metal cations.

The reactivity of both fresh, unaltered basaltic glass and crystalline basalt from the Stapafell mountain (southwestern Iceland) has been investigated (Table 1). The crystalline basalt was assumed to contain five mineral phases: 41 vol.% labradoritic plagioclase ( $\text{Ca}_{0.7}\text{Na}_{0.3}(\text{Fe}, \text{Mg})_{0.1}\text{Al}_{1.5}\text{Si}_{2.4}\text{O}_8$ ), 34 vol.% augitic pyroxene ((Ca, Mg, Fe, Ti) $_{1.9}\text{Al}_{2.5}\text{Si}_{1.8}\text{O}_6$ ), 16 vol.% olivine ( $\text{Mg}_{1.7}\text{Fe}_{0.3}\text{Si}_{1.0}\text{O}_4$ ), 5 vol.% opaque minerals (Fe-Ti oxide-hydroxides), 4 vol.% glass [20].

Table 1. Composition of crystalline and glassy basalt from the Stapafell Mountain, together with mid-ocean ridge basalt (MORB) reference. The data were obtained by XRF analysis and reported as weight percentage of oxides.

	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub> *	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	TiO <sub>2</sub>	MnO	P <sub>2</sub> O <sub>5</sub>	LOI	Total
Basaltic glass [11]	48.1	14.6	10.9	9.1	11.8	2.0	0.3	1.6	0.2	0.2		98.8
Crystalline basalt [20]	47.9	13.4	12.3	10.0	12.2	1.5	0.3	1.6	0.2	0.2	-0.5	99.1
MORB [21]	49.9	15.3	10.3	8.0	11.5	2.6	0.1	1.4	0.2	0.2		99.4

\* Most iron in the +2 form [11]

Despite their different crystallinity, both rocks exhibit bulk compositions similar to each other and to mid-ocean ridge basalts (Table 1). However, the reactivity of basalts decreases with increasing crystallinity (e.g. basaltic glass dissolves faster than crystalline basalt) [22-24]. Also, fresh basalt alters over time. Alteration consists of: i) the decomposition of primary basaltic phases, and ii) the precipitation of secondary minerals [25]. The common order of susceptibility to alteration for a basaltic rock is glass ~ olivine > plagioclase > pyroxene > opaque minerals [26], but pH, temperature, and mineral composition are all variables that influence the relative reactivity of these phases [27, 8]. For example, studies suggest that pyroxene will preferentially be dissolved at acidic to neutral pH conditions, releasing Mg and Fe, whereas plagioclase at basic pH with the release of Ca and Al [20, 29, 30].

The identity of the secondary minerals that form within a basalt depends on the thermal conditions and the initial rock composition [31] (Fig.1). At low temperature (<100 °C), glass, olivine, and plagioclase are commonly replaced by carbonates and clay minerals, and in some cases by quartz, zeolites, and sphene, whereas pyroxene is commonly replaced by clays. At temperature >200 °C, albite and chlorite become the predominant alteration phases, whereas at temperature exceeding ~220 °C epidote and actinolite tend to replace the original plagioclase and pyroxene, respectively. However, the chemistry of the fluids circulating through the rock can modify the average basalt solubility, and in turn the type of forming secondary minerals, such as for the case of seawater that induces the precipitation of mostly clay (smectite and saponite) at 250-300 °C [32, 33]. Pyrite and sphene precipitate over the whole temperature range as alteration products of the opaque minerals.

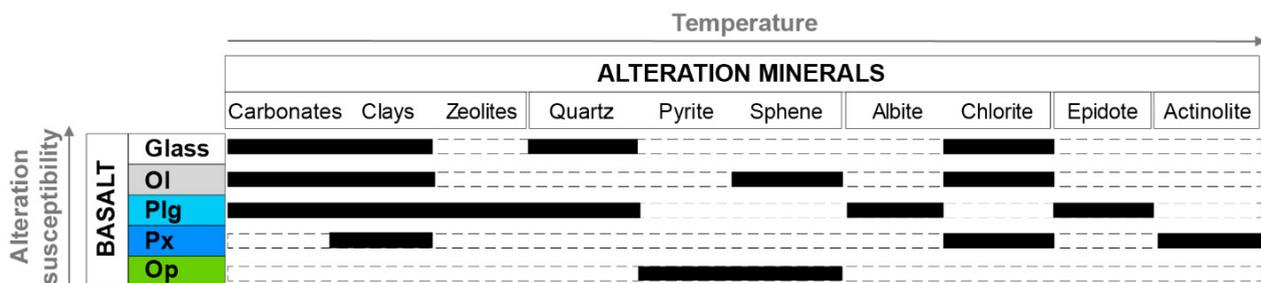


Fig. 1. Primary phases characterizing the crystalline basalt [20] and their common alteration minerals [31, 34], shown as a function of alteration susceptibility and temperature, respectively. Ol-Olivine; Plg-Plagioclase; Px-Pyroxene; Op-Opaque minerals.

Amongst the secondary minerals, clays (which commonly contain less than 1 wt.% Ca [35]), zeolites, chlorite, epidote, actinolite, and carbonates can be the sources of divalent cations for mineral carbonation. Work during the CarbFix CO<sub>2</sub> injections suggest that calcium is the most important divalent cation for basalt carbonation at temperatures of not more than 40 °C, leading rapidly to the precipitation of Ca-carbonates [18, 36]. To begin assessing the role of various alteration phases to provide this Ca we have compared the Ca release rates from basaltic glass, crystalline basalt, plagioclase, pyroxene, epidote, and actinolite.

### 3. Calcium release rates

Most of the dissolution rates of silicate rocks and minerals provided in the literature are based on silicon release, due to its position within the mineral framework [11]. Hence, the Ca release rates were calculated for basaltic glass (Na<sub>0.08</sub>Ca<sub>0.263</sub>Mg<sub>0.281</sub>Fe<sub>0.188</sub>Al<sub>0.358</sub>Si<sub>3.32</sub>), labradoritic plagioclase (Ca<sub>0.5</sub>Na<sub>0.5</sub>AlSi<sub>3</sub>O<sub>8</sub> [20, 37]), diopsidic pyroxene (Ca<sub>0.97</sub>Mg<sub>0.89</sub>Fe<sub>0.08</sub>Al<sub>0.04</sub>Si<sub>1.99</sub>O<sub>6</sub> [38]), and actinolite (Na<sub>0.41</sub>Mg<sub>3.10</sub>Ca<sub>2.70</sub>Fe<sub>0.97</sub>Al<sub>0.85</sub>Si<sub>7.38</sub>O<sub>22</sub>(OH)<sub>2</sub> [39]) assuming stoichiometric dissolution. Calcium release rates for crystalline basalt, and epidote [40, 41] were directly measured at steady-state (Fig. 2).

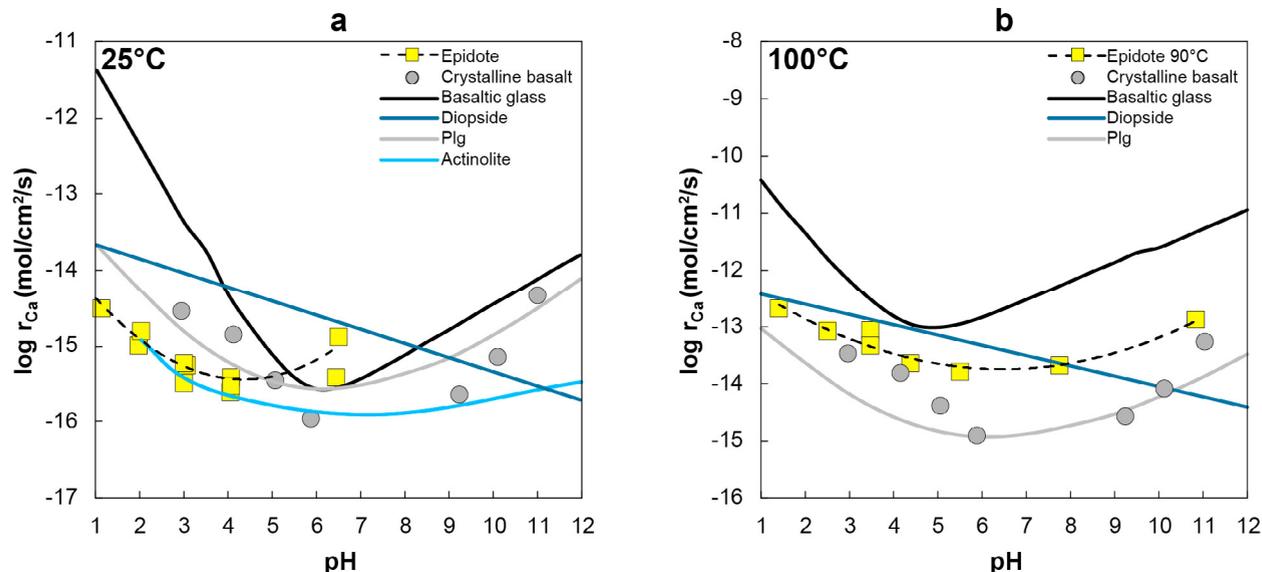


Fig. 2. Comparison of BET surface area normalized calcium release rates as a function of pH, at 25 (a) and 100 °C (b). Epidote at 25 °C [40] and 90 °C [41]; diopside [38], labradoritic plagioclase [20, 37], crystalline [20] and glassy [42] basalt at 25 and 100 °C; actinolite only at 25 °C [39].

All the release rates have been normalized to the Brunauer-Emmett-Teller (BET) surface area, and are provided for 25 and 100 °C. In some cases, the Si based dissolution rates at 100 °C were not available, and the rates for this

temperature have been extrapolated as described below. The Ca release rates for the labradoritic plagioclase have been extrapolated from 25 to 100 °C by applying a constant activation energy of 18.2 kJ/mol, consistent with that reported for anorthitic plagioclase [43]. Calcium release rates for the diopside were extrapolated from 70 to 100 °C by applying an activation energy of 37.7 kJ/mol [38]. For crystalline basalt, the Ca release rates at 100 °C were generated from rates reported at 25 °C [20] using a constant activation energy of 30 kJ/mol. Actinolite release rates have been found only for 25 °C [39].

Calcium release rates exhibit the common U-shaped variation as a function of pH for all minerals other than diopside, whose Ca release rate decreases continuously with increasing pH. At 25 °C, Ca release from crystalline and basaltic glass increase more rapidly with decreasing pH than for epidote, plagioclase, and pyroxene. At 25 °C and neutral pH conditions all the phases besides diopside exhibit similar BET surface area normalized Ca release rates; each are within  $\pm 0.5 \log \text{ mol/cm}^2/\text{s}$ . In contrast, at 100 °C crystalline basalt and plagioclase are estimated to have the same BET surface area normalized calcium release rates over the whole pH range, epidote dissolves slightly faster, and diopside becomes the most reactive amongst the crystalline phases only at the specific pH range from 2 to 7.

#### 4. Discussion and conclusions

Overall, basaltic glass appears to liberate Ca from 0.5 to 3.0 orders of magnitude faster than fresh and altered crystalline basalts. At 25 °C and neutral pH, all the rocks show comparable release rates, which correspond to the lowest reactivity conditions of most of these phases.

The close correspondence between the Ca release rate pH trends between fresh crystalline basalt and plagioclase shows that this mineral is the main source of Ca within the crystalline rock for Ca-carbonate formation at both 25 and 100 °C. Furthermore, results indicate that pyroxene is less reactive than Ca-rich plagioclase. On the other hand, the alteration of plagioclase to epidote appears to increase the BET surface area normalized Ca release rates of the rock, mainly at 100 °C.

These results suggest that, for the geological storage of CO<sub>2</sub>, an altered mafic reservoir can be as efficient as a fresh crystalline basaltic reservoir so long as these secondary phases do not substantially alter fluid flow pathways. Such conclusions would be improved by further insight into:

- the dissolution rates of secondary phases and bulk altered rocks;
- the effect of aqueous fluid composition, such as seawater compositions, on the overall phase reactivity [12], to account for potential offshore scenarios;
- the feasibility of CO<sub>2</sub> injection into altered basaltic reservoirs; notably the presence of clay minerals in these rocks may alter substantially their injectivity.

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