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EXPERIMENTS OF CO₂- BASALT- LIQUID INTERACTIONS AND GEOCHEMICAL MODELS: IMPLICATIONS FOR CO₂ MINERALIZATION IN BASALTS

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ABSTRACT

The rising amount of CO₂ in the atmosphere increased the average temperature of the Earth. The UN made the Paris Agreement in 2015 to significantly mitigate greenhouse gas (GHG) emissions and to try to limit the average global temperature increase to 2°C by the end of the current century. CO₂ constitutes a significant part of the GHG, and carbon capture and storage (CCS) is one of the most promising techniques to achieve these prefixed targets. Several previous studies focused on carbonation of natural sedimentary, mafic and ultramafic rocks, while detailed experimental studies on natural basalts systems are rare in the literature. Here, we carried out a detailed study on interactions between CO₂, liquid and alkali basalts from a young continental lava field in south Saudi Arabia (Al-Birk) coupling scanning electron microscopy (SEM), X-ray diffraction (XRD), X-ray fluorescence (XRF), Fourier-transform infrared spectroscopy (FTIR), and Raman spectroscopy. In parallel, liquid products were analyzed via inductively coupled plasma–optical emission spectrometry (ICP-OES). Laboratory experiments such as ageing tests were carried out through CO₂ injection in grinded basalts to unravel chemical reactions occurring during precipitation/dissolution processes at P-T-X. Eight ageing tests were carried out to evaluate the effects of different parameters on the precipitation of carbonates which are experiment time and fluid sampling method. Experimental observations were tested with geochemical modeling that was used to evaluate carbonate saturation and mineral stability under the investigated conditions. The absence of measurable carbonate precipitation under the investigated experimental conditions indicates that reaction time up to 552 h was insufficient to induce detectable mineral carbonation. Nonetheless, mineral dissolution was observed throughout the experiments, indicating ongoing interaction between CO₂-basalt-liquid and the release of divalent cations. Although these processes did not result in detectable carbonate formation, these findings highlight the importance of the initial alteration state and initial mineralogical composition of basalt in controlling dissolution pathways and the potential for subsequent CO₂ mineralization.

INTRODUCTION

Carbon dioxide (CO₂) levels in the atmosphere are increasing at an unprecedented rate in human history (IPCC,2023), likely because of a complex combination of combustion of fossil fuels for energy generation, deforestation, and agricultural activities. To limit Earth's temperature increase to <2 °C by the end of the 21st century, the International Panel on Climate Change (IPCC) reported that

Greenhouse gas (GHG) emissions should be reduced by 45-50% by 2050, with constant reductions during the rest of the 21st century (Kabir et al., 2023; Olhoff, 2025).

In this context, CCS technology is a possible solution to decrease global CO₂ levels by diminishing its emission in the atmosphere (e.g. Xu et al., 2019; Mitchell et al., 2010). This technology is based on separation process of CO₂ from high energy consuming industrial or energy production-related energy sources followed by transportation and storage in appropriate subsurface reservoirs (Novak Mavar et al., 2021) such as sedimentary basins (Gaus, 2010; Pokrovsky et al., 2009; Shi et al., 2013), basaltic or ultramafic rocks (Oelkers et al., 2026; Omar et al., 2025; Rigopoulos et al., 2018), deep saline aquifers (Giammar et al., 2005, Mim et al., 2023) and oil and gas reservoirs (Li et al., 2021, Ciotta et al., 2021). Depending on reservoir conditions, dissolved CO₂ may subsequently react with host-rock minerals to form stable carbonate minerals (mineral trapping, Cao et al., 2024).

Typical rock formations suitable for CO₂ mineralization should be able to transport fluids and react efficiently with CO₂; hence, high porosity, high permeability and a mineralogy able to be transformed to stable carbonate minerals are all fundamental characteristics. The Carbfix project demonstrated that over 95% of the CO₂ injected within basaltic rocks at Hellisheidi SW (Iceland) was mineralized to carbonate minerals in less than two years (Matter et al., 2016). The injection of CO₂-water mixture accelerates reactions between mafic rocks and CO₂ because of higher temperature and pressure in reservoir conditions (Snæbjörnsdóttir et al., 2020). Since CO₂ is dissolved in water, HCO₃⁻ and H⁺ ions will be formed with consequent decrease in initial pH. Extent of this reaction depends on CO₂ partial pressure, temperature and solution salinity. The initial pH drop promotes the dissolution of Ca-Mg-Fe silicate phases (e.g. olivine, plagioclase, and clinopyroxene), consuming protons (H⁺) and releasing calcium, magnesium and iron cations into solution. This reaction gradually increases pH, favoring carbonate precipitation. (Zhang and DePaolo, 2017; Gudbrandsson et al., 2011; Gislason et al., 2010). As the solution pH rises, the dissolved cations react with CO₃²⁻ to form stable carbonate minerals. This step is strongly controlled by the dissolution rate of basalt minerals and pH conditions. Thanks to suitable geochemistry of these reactions and success of this pilot project, further research on CO₂ mineralization in basaltic rocks were carried out to implement in-situ mineralization and storage worldwide (Cao et al., 2024).

Laboratory investigations of basalt carbonation are commonly conducted using either batch reactors or flow-through experimental systems. Batch experiments are particularly useful for studying long-term CO₂-basalt-liquid interactions and identifying reaction products, whereas flow-through systems enable the investigation of fluid circulation effects and reaction kinetics. In both configurations, an external CO₂ source is typically used to maintain the desired pressure conditions (Gysi and Stefansson, 2012; Wells et al., 2017).

Many experimental studies have investigated CO₂-basalt-water systems under a range of temperature and pressure conditions (e.g., Lu et al., 2024; Xiong et al., 2018; Wells et al., 2017). A study of carbonation of Jizan basalts at 25 and 100 °C, indicated that more than 95% of injected CO₂ should be mineralized in basaltic rocks within 100 years at 25 °C, and 5 years at 100 °C (Oelker et al., 2022). Similarly, high-pressure batch experiments between 50 and 200 °C at 300 bar showed net CO₂ sequestration of approximately 8% and 26% by mass in tholeiitic basalt and picrite, respectively, with magnesite as the dominant carbonate phase (Rosenbauer et al., 2012).

Despite substantial progress, significant uncertainties remain. Batch reactions are conducted in static autoclaves that neglect the transport aspect. When CO₂ is injected into the subsurface, the solution will move through pores providing CO₂ storage and flow pathway for fluid and carbonation (Liu et al., 2019). Therefore, integrating experiments with reactive-transport modeling—using tools such as TOUGHREACT (Xu et al., 2006) and PHREEQC (Parkhurst and Appelo, 1999)—is essential to predict long-term behavior and optimize injection strategies.

The aim of this work is to investigate CO₂-basalt-liquid interactions through laboratory experiments and geochemical models to assess CO₂ sequestration potential in basaltic formations under the controlled conditions (experimental duration and fluid-sampling method) and to evaluate the possible challenges associated with CO₂ storage. Eight ageing experiment tests are conducted in a micro

reactor mimicking the subsurface condition to understand the effectiveness of carbonation reactions in basalts. Batch experiments were conducted on altered alkali basalts from Saudi Arabia. Grinded basalt samples were reacted with CO₂ injected water in a pressurized autoclave under controlled temperature and pressure conditions for reaction times ranging from 2 h to 552 h. The mineralogical and fluid composition of both the starting material and experimental products and of the liquid phases were characterized combining XRD, XRF, SEM, FTIR, Raman Spectroscopy, and ICP-OES. Geochemical modeling was used as a supporting tool to interpret fluid chemistry and evaluate carbonate saturation under the investigated experimental conditions. We show that experiments promote progressive dissolution of altered basalt leading to significant changes in fluid chemistry and solids over reaction times ranging from 2 h to 552 h.

GEOLOGICAL BACKGROUND

One alkali basalt was selected from a lava field outcropping in southern Saudi Arabia. The sample was selected from a suite of Quaternary basalts from the Al Birk lava field, at ~100 km south of the city of Al Birk (Fig. 1). This volcanic field forms an irregular elongate area covering about 1800 km² (Coleman et al., 1983). The lava flows consist primarily of olivine bearing alkaline basalts and frequently contain mantle xenoliths and are also associated with less voluminous pyroclastic deposits (Sanfilippo et al., 2019). The rock is a porphyritic alkali basalt containing large olivine (Ol) and plagioclase (Pl) phenocrysts ranging in size from 0.5 to 4 mm (Fig. 2). Olivine phenocrysts have high forsterite (up to 92 mol%) contents, although in exceptional cases they show normal zoning to more fayalitic compositions (Fo 20-26 mol%; Sanfilippo et al., 2024). Plagioclase phenocrysts have anorthite-rich cores (An 53-70 mol%) surrounded by rims with more albitic composition (28-46 mol%). The phenocrysts are also associated to mantle xenoliths made up of Ol and orthopyroxene (Opx) and, more rarely, chromian spinel (Fig 2e). The groundmass is crypto- to micro-crystalline and consists of euhedral to subhedral plagioclase laths (10-50 microns) and olivine (25-500 microns) embedded in subophitic clinopyroxene. A partly altered glassy groundmass is also visible as interstitial phase, less than 50 microns in size. Selected sample also includes secondary minerals replacing the magmatic phases: olivine is partially replaced by bowlingite-like aggregates (i.e., smectite, chlorite, serpentine), plagioclase is rarely altered into microaggregates of albite-epidote whereas clinopyroxene in the groundmass is generally fresh. The original glass is converted into clay minerals. Small, parallel veins and microcracks (5-20 microns) filled by calcite or clay minerals crosscut the sample.

Sample DIS14-506 was selected for the ageing experiments because petrographic observations and whole-rock geochemical analyses indicated a relatively high abundance of olivine and elevated MgO (10.13 wt.%), FeO (8.95 wt%) and CaO (10.03 wt%) in whole rock, compared to the other collected samples. These characteristics make the sample particularly suitable for investigating CO₂ mineralization processes, as olivine and Ca-pyroxenes represent important sources of divalent cations required for carbonate precipitations. Although the experiments were conducted on a single sample, its mineralogical assemblage (olivine, plagioclase, and clinopyroxene) is characteristic of many alkaline basalts occurring within the Al Birk volcanic field.

METHODOLOGY

The methodology of this study integrates laboratory ageing experiments with geochemical modeling to investigate CO₂-basalt-liquid interactions. Mineralogical and chemical characterization of both starting materials and post-experiment products was carried out using SEM, XRD, XRF, FTIR, and Raman spectroscopy. These analytical techniques are briefly introduced in this chapter, with detailed

descriptions provided in the Supplementary Materials. The experimental setup and geochemical modeling are described below.

Sample Preparation

The sample was initially broken into smaller fragments using a geological hammer, then dry-sieved into grain-size fraction of 125–250 μm , the latter being the primary size fraction used in the experiments.

Particle-size distribution for the 125–250 μm fraction was measured using a Mastersizer 3000 using image-analysis techniques. To remove fine particles adhering to grain surfaces, samples used in some experiments were washed with ethanol, followed by repeated particle-size analyses to verify whether washing had effectively improved granulometric uniformity. A portion of the material was further powdered for XRD analysis.

Experimental Setup

The experimental matrix (Fig. 3) was developed to investigate the influence of experimental duration and fluid-sampling method. Other minor experimental parameters were changed during the research activity (grain size, and sample washing or non-washing before the experiment) but here we present a homogeneous experiment set in order to make the results comparable.

The temperature and pressure conditions were selected based on previous experimental studies (Van Pham et al., 2012; Adeoye et al., 2017; Schaef and McGray, 2009; Xiong et al., 2018; Menefee et al., 2018; Cao et al., 2024) and on preliminary geochemical simulations performed in this study using PHREEQC (Parkhurst and Appelo, 1999). The selected conditions (110 °C and 40-50 bar) were intended to represent elevated-temperature subsurface environments relevant to geological CO₂ storage in basaltic rocks, while also promoting measurable CO₂-basalt-liquid reactions within laboratory timescales. Preliminary simulations explored the effects of temperature and liquid salinity on calcite solubility and CO₂ behavior in CO₂-basalt-liquid systems (Fig. 4). The results indicated that calcite solubility reaches a minimum between approximately 100 and 150 °C, supporting the selection of 110 °C as the experimental temperature. These simulations were used as a preliminary tool to guide the design of the ageing experiments and are not intended to reproduce the full complexity of the experimental system. Milli Q water was selected as the reaction fluid to provide a well-defined initial fluid composition, minimize background ionic influences and facilitate interpretation of CO₂-basalt-liquid interaction during the experiments (Kikuchi et al., 2023; Pan et al., 2025).

A blank experiment was carried out prior to the experiments. It lasted 1 h under identical temperature and pressure conditions in the absence of basalt in order to obtain the starting concentration of various species in the liquid. In each experiment, one or more parameters were varied to investigate their effect on carbonate precipitation. Experiments were performed at 110 °C, CO₂ pressure variable from (40 to 50 bar). The selected temperature falls within the range reported to promote carbonation reactions (Adam et al., 2013; Kumar et al., 2017; Xiong et al., 2017; Menefee et al., 2018; Voigt et al., 2021) while applied pressure ensure sufficient CO₂ dissolution in the fluid phase. A water-to-rock ratio of 10 (2 ± 0.01 gr basalt powder (125-250 μm) + 20 mL Milli-Q water) was used, with continuous stirring to make sure to have a homogeneous mixture throughout the experiments. According to Delerce et al. 2023, element release rates from altered basalt peaks at 120 °C, and carbonation can occur rapidly at temperatures above 100 °C, whereas mineral carbonation is slower at lower temperatures. The chosen water-to-rock ratio was decided following previous batch carbonation studies (e.g., Rosenbauer et al., 2012). This ratio provides sufficient fluid volume for post-experiment chemical analyses while maintaining effective water-to-rock ratios, it offers a controlled and reproducible experiment system for investigating dissolution processes and the potential for carbonate formation.

The experiment program (Table 1) was organized into three series, each series was designed to investigate basalt mineral dissolution and possible carbonate mineral precipitation.

In a first series of experiments (EXP-368, 370, 371), three experiments have been carried out with different durations (2-16-48h) to determine the optimal reaction time. Experiments are conducted maintaining constant experimental conditions (110 °C, 40-50 bar CO₂ pressure, grain size of 125-250 µm, unwashed material, and in-situ sampling). This initial series established the baseline evolution of fluid chemistry and mineralogical changes over time.

Then, based on the results of first series, in subsequent run (EXP-372), we kept experiment time fixed at 48 h. We varied the sampling method (in-situ or at the end of experiment) aiming to enhance solid reactivity. Two different sampling methods were considered. In-situ sampling fluids are sampled right after the end of experiment, before cooling. On the contrary, the other method consists in the collection of experiment fluids after about 30 minutes from the end of experiment at ambient T and after degassing the reactor. This process could increase pH of the solution, and this can favor carbonate precipitation.

For the final series (EXP-378, 379, 386), we prolonged the experiment duration to 168 h (96 h at run temperature and pressure, then 72 h with reactor cooled and degassed) for EXP-378 and 504 h (336 h constant pressure, then 168 h reactor degassed) for EXP-379 and finally experiment duration for 552 h (144 h constant pressure, then 408 h with reactor degassed) for EXP-386 (552 h). This series was designed to evaluate whether extended reaction times and post-reaction cooling conditions promoted detectable carbonate precipitation after the initial basalt dissolution stage. All experiments were performed with Milli-Q water and CO₂ at 110 °C under an initial CO₂ pressure of 40-50 bar using a water-to-rock mass ratio (W/R) of 10:1 and a basalt grain size of 125-250 µm. In EXP-378, EXP-379, and EXP-386, the starting basalt was washed with ethanol prior to the experiment, and liquid sampling was carried out at room temperature after depressurization. Stirring was applied only during the high-pressure stage of the experiments.

Geochemical Modeling of CO₂ Degassing

A simplified geochemical model was developed using PHREEQC (Parkhurst and Appelo, 1999) to evaluate the influence of CO₂ pressure on fluid pH during the depressurization stage of the ageing experiments (Fig. 5). The objective of the model was not to reproduce the complete basalt-water-CO₂ system but rather to isolate the effect of CO₂ degassing on solution acidity.

Simulations were performed at 110 °C using Milli-Q water equilibrated with a pure CO₂ gas phase. CO₂ pressure was progressively varied from experimental conditions (45 atm) to near-atmospheric values in order to simulate reactor depressurization after the experiments. The calculations were performed assuming thermodynamic equilibrium between the aqueous and gas phases.

The model intentionally excluded basalt dissolution and secondary mineral precipitation reactions. Therefore, buffering effects associated with silicate dissolution were not considered. The simulations should consequently be interpreted as a first-order approximation of pH evolution during degassing rather than a complete representation of the experimental system.

Model outputs included pH and total dissolved inorganic carbon concentration (C_{tot}), which were subsequently compared with experimental observations to evaluate the potential role of degassing carbonate precipitation.

RESULTS

Starting Material Characterization

The starting basalt sample was characterized using SEM-EDS, XRD, XRF, FTIR, and Raman Spectrometry to determine its mineralogical, chemical, and textural properties.

XRF analysis indicates the starting basalt (Table 2), has a typical basaltic composition with major oxides as follows: SiO₂ (44.54 wt.%) CaO (10.03 wt %), MgO (10.13 wt %), and FeO (T) (8.38 wt %), Na₂O (3.38 wt.%), K₂O (1.49 wt.%).

XRD analysis (<10 μm fraction) identifies Montmorillonite (7.2 wt.%), Diopside (32.9 wt.%), Nepheline (9.5 wt. %), Anorthite (34.7 wt.%), Forsterite (8.4 wt. %) and Ferrosilite (7.3 wt.%) as the main phases (Fig. 6). No carbonate phases were detected by XRD.

FTIR spectra display characteristic Si-O stretching bands at $\sim 1000\text{--}1100\text{ cm}^{-1}$, consistent with primary silicate minerals, and no distinct carbonate absorption bands (Fig. 7). Raman spectrometry indicates carbonate phases at 9 of the 560 measurement points analyzed in the starting basalt, corresponding to a carbonate detection frequency of 1.61%. Of these carbonate-positive spectra, seven were assigned to calcite and two to dolomite (Supplementary Material).

SEM BSE images and corresponding EDS elemental maps (Si, Al, Mg, and Ca) of the starting basalt (DIS14-506), revealed the coexistence of relatively fresh and altered basaltic glass (Fig. 8).

Experimental Products Characterization

Solid Phases

Solid products were analyzed using SEM-EDS, SEM imaging, XRD, FTIR, Raman Spectroscopy, and EPMA to evaluate mineralogical and textural changes relative to the starting material.

SEM imaging of short-duration experiments (2-168 h) shows no visible differences compared to the starting material (Fig. 9). No dissolution textures or newly formed mineral phases were observed. In non-washed samples, micrograins partially obscure grain surfaces. In longer experiments (504-552 h), localized dissolution features and alteration zones are visible (Fig. 10). No distinct carbonate minerals were identified.

Bulk XRF analyses of the post-experiment analyses samples (EXP-378 (168 h), EXP-379 (504 h), and EXP-386 (552 h)) show only minor variations in major oxide composition relative to the starting basalt (Table 2). SiO_2 , CaO, and FeO (T) remain broadly constant throughout the experiments, while slight decreases in Al_2O_3 and K_2O and relatively higher MgO and Na_2O contents are observed in the reacted samples. Overall, these limited variations indicate that the ageing experiments did not substantially modify the bulk chemical composition of the basalt, despite evidence of localized mineral dissolution from SEM observations and liquid analyses results.

Quantitative XRD analyses show the bulk mineral assemblage remained largely preserved throughout the ageing experiments (Fig. 6). Anorthite remained the dominant mineral phase (28.6-38.1 wt.%), while pyroxene was represented by diopside and augite. Compared to initial basalt, diopside generally decreased whereas augite became progressively more abundant, showing compositional variation within pyroxenes. Dolomite was detected in EXP-378 (168 h) (1.2 wt. %) and in EXP-386 (552 h) (0.1 wt.%), whereas it was not identified in EXP-379 (Fig. 6).

FTIR spectra show a progressive decrease in the intensity of Si-O stretching bands ($\sim 1000\text{--}1100\text{ cm}^{-1}$) with increasing reaction time (Fig. 7). No distinct carbonate absorption bands were detected. Raman spectrometry identifies carbonate phases at 8 of the 632 measurement points analyzed in EXP-386, corresponding to a carbonate detection frequency of 1.27 %. Seven of these spectra were assigned to dolomite and one to calcite (Supplementary Material).

Liquid Phases

ICP-OES analysis of post-experiment fluids reveal time-dependent variations in dissolved cation concentrations (Fig. 11). In short-duration experiments (2-48 h), relatively elevated cation concentrations were measured. At 2h, Ca reached 3.64 mM, Mg 1.56 mM, Na 2.22 mM, and K 1.05 mM. In contrast, the experiment conducted without CO_2 at 2h (EXP-369) showed significantly lower concentrations (Ca: 0.16 mM; Mg: 0.01 mM). At 16 h and 48 h, Ca concentrations increased to 4.24 and 4.01 mM, while Mg concentrations were 2.26 and 2.43 mM respectively. Also, Na and K concentrations increased progressively with time. In EXP-377,378 (168 h), Ca reached 5.49 and 5.19 mM, while Mg, Na, and K remained relatively stable. Experiments conducted with coarser grain size showed lower dissolved concentrations.

In long-duration experiments (504–552 h), Na concentrations increased further (10.7 and 8.7 mM), whereas Ca decreased to 1.99 and 1.65 mM and Mg decreased to ~0.29 mM.

DISCUSSION

Post-ageing analyses suggest progressive basalt dissolution during the ageing experiments, whereas evidence for carbonate precipitation remained limited. Although carbonate-bearing phases were identified after the experiments, their origin cannot be unequivocally established because carbonate was also present in the starting material. Progressive dissolution textures became more apparent with increasing reaction duration, while ICP-OES analyses demonstrated continued release of dissolved cations. These results suggest that factors such as reaction duration, depressurization, and the mineralogical characteristics of the starting material influenced the overall reactivity of the system.

Influence of Starting Material Composition

The efficiency of basalt for CO₂ mineralization is fundamentally controlled by its mineralogical composition. Primary minerals such as olivine, pyroxene, plagioclase and basaltic glass act as sources of cations and contribute to pH buffering during dissolution (Seifritz, 1990). The abundance, preservation, and alteration state of these minerals consequently influence the progress of liquid-basalt interaction during the ageing experiments.

Petrographic observations (Fig. 2), together with SEM-BSE imaging and elemental mapping (Fig. 8), demonstrate that the starting basalt used in this study had previously undergone localized fluid-mediated alteration. Two main alteration styles were identified. The first consists of partial alteration of basaltic glass, whereas the second corresponds to serpentinization of olivine along fractures and grain boundaries. Elemental maps further reveal localized Mg depletion adjacent to alteration fronts accompanied by the development of Al-bearing secondary phases. These observations indicate that part of the original reactive mineral assemblage had already been transformed into secondary alteration products before the experiments, reducing the proportion of fresh mineral surfaces directly exposed to the reacting fluid.

Previous studies have demonstrated that the degree of alteration strongly influences basalt dissolution kinetics. Delerce et al., 2023 showed that altered basalts release Si, Mg, and Ca at rates one to three orders of magnitude lower than fresh basalts under comparable conditions. They attributed this behavior to the loss of highly reactive primary mineral surfaces through pre-existing alteration. Nevertheless, increasing temperature substantially enhances dissolution kinetics, and altered basalts reacted at approximately 120 °C exhibiting elemental release rates comparable to those measured for fresh basalts at ambient temperature. Likewise, experimental studies have shown that olivine dissolves relatively rapidly, supplying Mg and Fe during the early stages of reaction, whereas pyroxene dissolves more slowly and progressively contributes Ca, Mg, and Fe over longer reaction times (Wells et al., 2017). The results obtained in the present study are consistent with progressive dissolution of basaltic minerals throughout the experiments. SEM observations reveal localized dissolution textures that become increasingly developed during the longest experiments (EXP-378, 379, and 386), while FTIR spectra indicate progressive modification of silicate minerals. ICP-OES analyses further demonstrate continuous release of major cations into solution, confirming ongoing CO₂-basalt-liquid interaction. Elevated Mg concentrations are consistent with dissolution of Mg-bearing phases, particularly olivine, although contributions from pre-existing alteration products cannot be excluded. Similarly, the high Ca concentrations are consistent with dissolution of Ca-bearing minerals such as plagioclase and pyroxene. Previous investigations of altered basalts have also reported dissolution of both primary silicate minerals and secondary smectitic alteration products during CO₂-basalt-liquid interaction, resulting in the release of Mg, Ca, and Na into solution (Wang et al., 2024). Although the present data do not allow the relative contribution of individual mineral

phases to be quantified, the combined mineralogical and geochemical observations consistently indicate progressive basalt dissolution during the experiments.

Despite this measurable dissolution and continued release of divalent cations, extensive carbonate precipitation was not detected. This indicates that cation release alone was insufficient to promote significant carbonate formation under the investigated experimental conditions. Comparison with previous studies suggests that pre-existing alteration of the starting basalt may have influenced the availability of reactive mineral surfaces and the evolution of fluid chemistry during the experiments. However, the extent to which alteration alone limited carbonate precipitation cannot be determined unequivocally from the present data, as carbonate formation is also controlled by additional factors including reaction kinetics, solution chemistry, and the evolution of CO₂ saturation during the experiments.

Effect of Degassing

Carbonate precipitation during CO₂-basalt-liquid interaction is controlled by the coupled evolution of dissolved inorganic carbon, solution chemistry, and mineral dissolution. Injection of CO₂ initially lowers the pH through carbonic acid formation, promoting dissolution of primary basaltic minerals and the release of divalent cations into solution. As dissolution progresses, proton consumption during silicate weathering progressively buffers the solution, favoring conditions that may eventually lead to carbonate supersaturation (Gysi and Stefansson, 2012).

Several experimental studies have shown that dissolved Ca and Mg concentrations initially increase as basalt dissolves and subsequently decrease or stabilize once secondary mineral precipitation begins (Rosenbauer et al., 2012; Oelkers et al., 2012; Oelkers et al., 2022). In the present study, ICP-OES analyses likewise demonstrate progressive release of major cations during the experiments. However, unlike these previous studies, extensive carbonate precipitation was not detected despite prolonged reaction times. This suggests that cation release alone was insufficient to promote significant carbonate formation under the investigated experimental conditions.

To investigate whether depressurization could favor conditions for carbonate precipitation, a conceptual geochemical model was developed using PHREEQC (Fig. 5). The model simulates the effect of decreasing CO₂ pressure on solution chemistry in a CO₂-liquid system and predicts that progressive CO₂ degassing increases solution pH by shifting the carbonate equilibrium toward lower dissolved CO₂ concentrations. It should be emphasized, however, that the model does not include basalt dissolution or the buffering effect of mineral reactions and therefore represents only the evolution of the fluid phase.

Based on this conceptual model, an extended low-pressure stage was incorporated into the longest experiments (EXP-378 (168 h), 379 (504 h), and 386 (552 h)) to allow the system to evolve after depressurization. The rationale was that prolonged residence at reduced CO₂ pressure might favor conditions more conducive to carbonate precipitation. Nevertheless, because pH was not measured during the experiments, it cannot be verified whether the predicted increase in pH actually occurred. Consequently, the effect of depressurization on carbonate saturation remains uncertain.

Previous studies have similarly suggested that CO₂ degassing may promote carbonate precipitation by reducing dissolved CO₂ and increasing carbonate ion activity. For example, Cosmo et al., 2019 reported that degassing accounted for a substantial proportion of calcite precipitation, building upon mechanisms previously described by Herman and Lorah, 1987, Dreybrodt et al., 1992, and Papadimitriou et al., 2004. However, the effectiveness of degassing depends on the combined evolution of fluid chemistry, cation availability, and mineral reaction kinetics. The results of the present study indicate that depressurization alone was insufficient to induce extensive carbonate precipitation, suggesting that additional controls- including the mineralogical characteristics of the starting basalt and the kinetics of mineral dissolution- also influenced the overall carbonation process.

Effect of Experiment Duration

Reaction time plays a critical role in controlling the extent of basalt-liquid interaction during CO₂ mineralization experiments. In short-duration experiments (2-168 h), SEM observations did not reveal detectable dissolution textures, although ICP-OES analyses showed progressively increasing concentration of dissolved major elements with increasing reaction time. This indicates that mineral dissolution had already begun during the early stages of the experiments but had not yet produced textural modifications detectable by SEM.

In contrast, the longest experiments, 379 (504 h), and 386 (552 h) exhibited localized alteration zones, indicating that prolonged reaction time promoted more extensive mineral-liquid interaction. The progressive development of dissolution features is consistent with continued silicate weathering under CO₂-rich conditions and agrees with the increasing degree of alteration observed in the reacted basalt grains.

The evolution of fluid chemistry also changed with reaction time. Ca and Mg concentrations increased during the early and intermediate stages of the experiments suggesting that these cations were progressively removed from solution but decreased during the longest experiments, whereas Na and K is consistent with ongoing dissolution of feldspar phases. Although the present data do not allow their fate to be determined unequivocally, the observed trends may reflect incorporation into secondary alteration minerals, limited carbonate precipitation below the detection limits of the analytical techniques or progressive evolution of the fluid toward basalt-water equilibrium.

The extended low-pressure stage introduced during the longest experiments may also have influenced fluid evolution during the later stages of reaction. However, because pH was not measured during the experiments, its effect on carbonate saturation cannot be assessed directly. Conceptual geochemical simulations performed with PHREEQC suggest that CO₂ degassing following depressurization may increase solution pH, but this interpretation remains qualitative because mineral buffering reactions were not included in the model and no experimental pH measurements were available.

Comparison with previous batch experiments shows that reaction time alone does not control carbonate precipitation. Several studies reported carbonate formation (e.g., Hovelmann et al., 2012; Kumar et al., 2017; Menefee et al., 2018); it may be although stressed that in these studies the experimental parameters were rather different (e.g. P_{CO2}, grain size, duration, amount of starting materials) from those used in this study. Similarly, despite experiments lasting up 552 h in the present study, only progressive basalt dissolution was observed, while carbonate precipitation remained limited. These comparisons suggest that reaction duration enhances basalt dissolution, but it is not, by itself, sufficient to induce mineral carbonation. Instead, carbonation appears to depend on the combined influence of factors including mineralogical composition, fluid chemistry, P-T conditions, and the evolution of water-rock interactions (Table 3).

Overall, the results indicate that increasing reaction duration enhanced basalt dissolution, as evidenced by both progressive development of dissolution textures and continued release of dissolved elements into solution. Nevertheless, prolonged reaction time alone was insufficient to generate extensive carbonate precipitation under the investigated experimental conditions. These findings indicate that carbonate mineralization was controlled not only by reaction time but also by the coupled effects of mineralogical composition, fluid evolution, and reaction kinetics.

CONCLUSIONS

To assess CO₂ mineralization potential of basalts in the Al Birk volcanic field, seven experiments were conducted using an altered basalt sample exploring the effect of different parameters that could affect the possibility of mineralization (Table 1). In each experiment, one or more parameters were changed to study their effect on carbonate precipitation. Experimental parameters were programmed after an evaluation of geochemical models such as PHREEQC to use the best T, P, and salinity conditions in experiments. The investigated parameters were mainly experimental time, sample pre-

treatment with ethanol washing, grain size, and controlled CO₂ degassing to reduce CO₂ pressure to evaluate the effect of lower acidity conditions on carbonate precipitation. The main conclusions are:

- 1) Reaction time appears to be the most important parameter influencing dissolution kinetics in the system. Up to 168 hours, the Ca concentration in the fluid increased, indicating ongoing mineral dissolution. However, in longer experiments (EXP-379 (504 h) and EXP-386 (552 h)), Ca content in water analysis decreased despite increasing reaction time. This could indicate calcite precipitation, but mineralogical analysis did not highlight calcite precipitation.
- 2) Minor operational procedural corrections (washing samples and the use or not of a stirrer during the experiments) apparently did not have a strong influence on the carbonate precipitation.
- 3) As expected, coarser grain size decreases the system reactivity due to minor reaction surface.
- 4) The fact that basalts coming from Al Birk are altered basalts, makes them less reactive with CO₂ with respect to fresh basalts. We tried to overcome this drawback degassing.

Further investigations are needed to fully understand the kinetics governing CO₂-basalt interactions, particularly the interplay between mineral dissolution, pH evolution, and carbonate precipitation under reservoir-relevant conditions. A comprehensive approach integrating laboratory experiments, geochemical modeling, and reactive transport simulations is therefore recommended to better constrain long-term carbonation processes at specific reservoir scale in order to reduce uncertainties and risks related to CCS activities.

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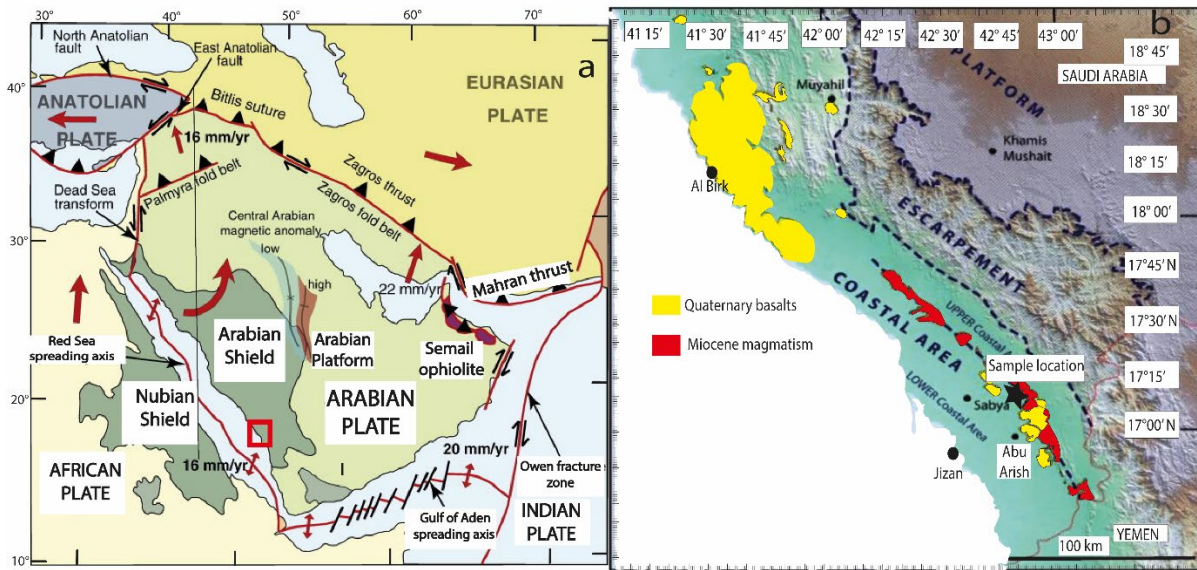


Fig.1 - (a) Tectonic map of Arabian plate and surrounding areas (Stern and Johnson, 2010); (b) Map of Al Birk volcanic field area, location in the red box in fig. 1a) (Mukhopadhyay et al., 2019). The yellow areas represent Quaternary aged basalts, and the orange areas represent Miocene magmatism. The star indicates the location of the studied sample. A colored version of the picture is available online.

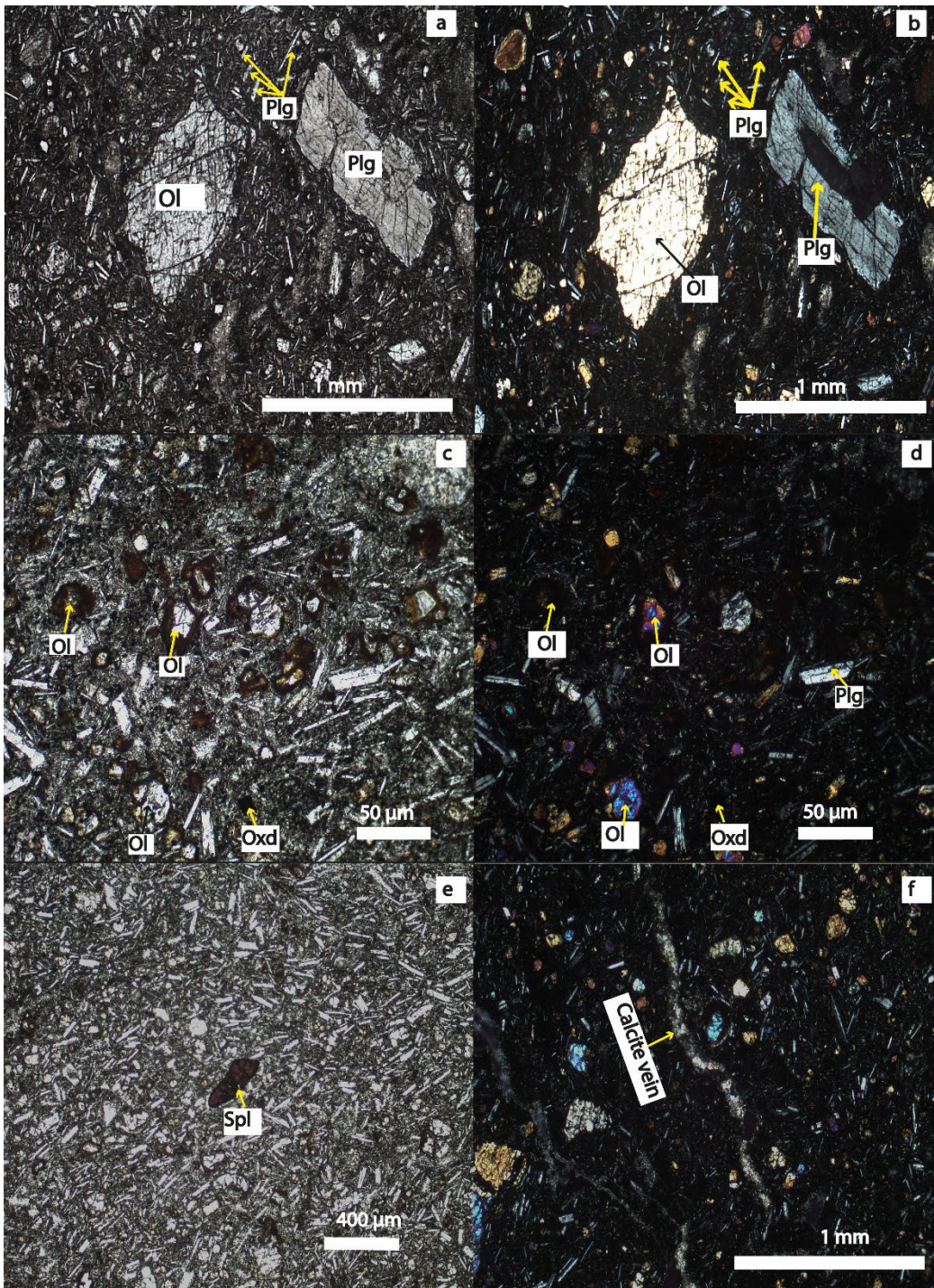


Fig. 2 – Petrographical images of Dis14-506 sample. Images taken using Optical Microscope. (a) Olivine and plagioclase phenocrysts with plagioclase microliths in crossed-polarized light (b) Olivine and plagioclase phenocrysts with plagioclase microliths in plane-polarized light (c) partially serpentinized olivine with plagioclase microliths in crossed-polarized light (d) partially serpentinized olivine with plagioclase microliths in plane-polarized light (e.) spinel phenocryst with plagioclase microliths in plane-polarized light (f) altered volcanic glass with small subparallel calcite vein and associated microcracks in crossed-polarized light. A colored version of the picture is available online.

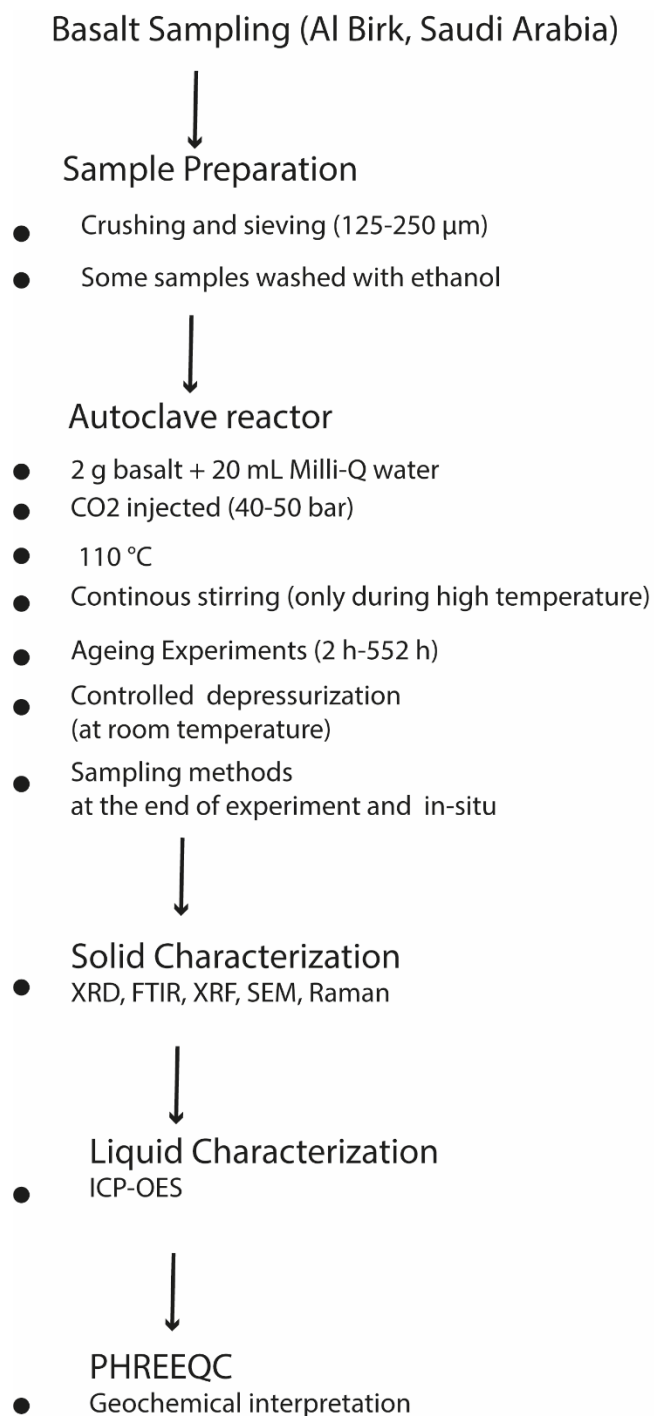


Fig.3 - A summary model to illustrate the experimental setup used in the laboratory. A colored version of the picture is available online.

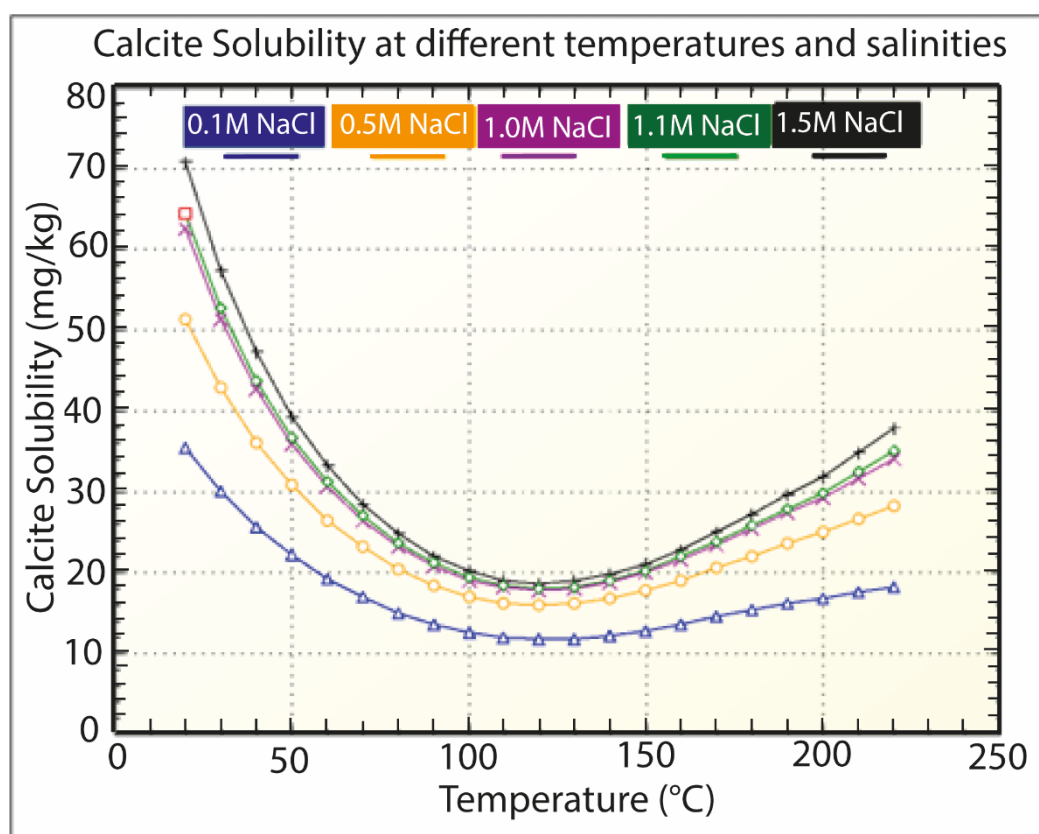


Fig. 4 - The geochemical model carried out using Phreeqc, graph showing the calculated calcite solubility in mg kg^{-1} in terms of increasing temperature (x axis) and different molarity of NaCl solutions (y axis). A colored version of the picture is available online.

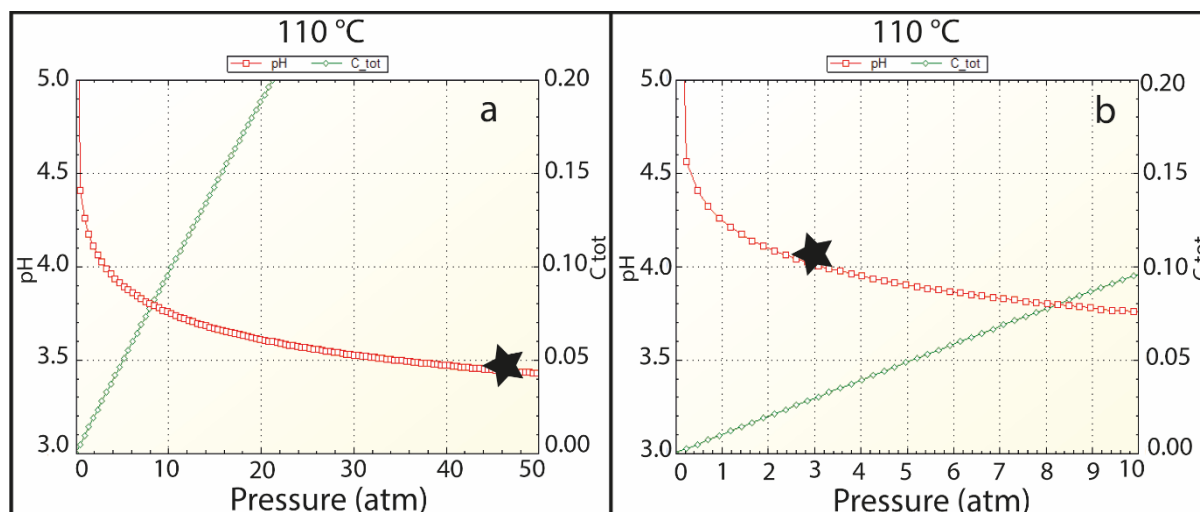


Fig. 5- a) The geochemical model carried out using Phreeqc, red line indicates pH values and green line indicates total Carbon in the liquid. (a) Studied basalt with CO₂ in the high-pressure environment (b) Studied basalt with CO₂ in the low-pressure environment. Stars indicate pressure values used in the experiments. A colored version of the picture is available online.

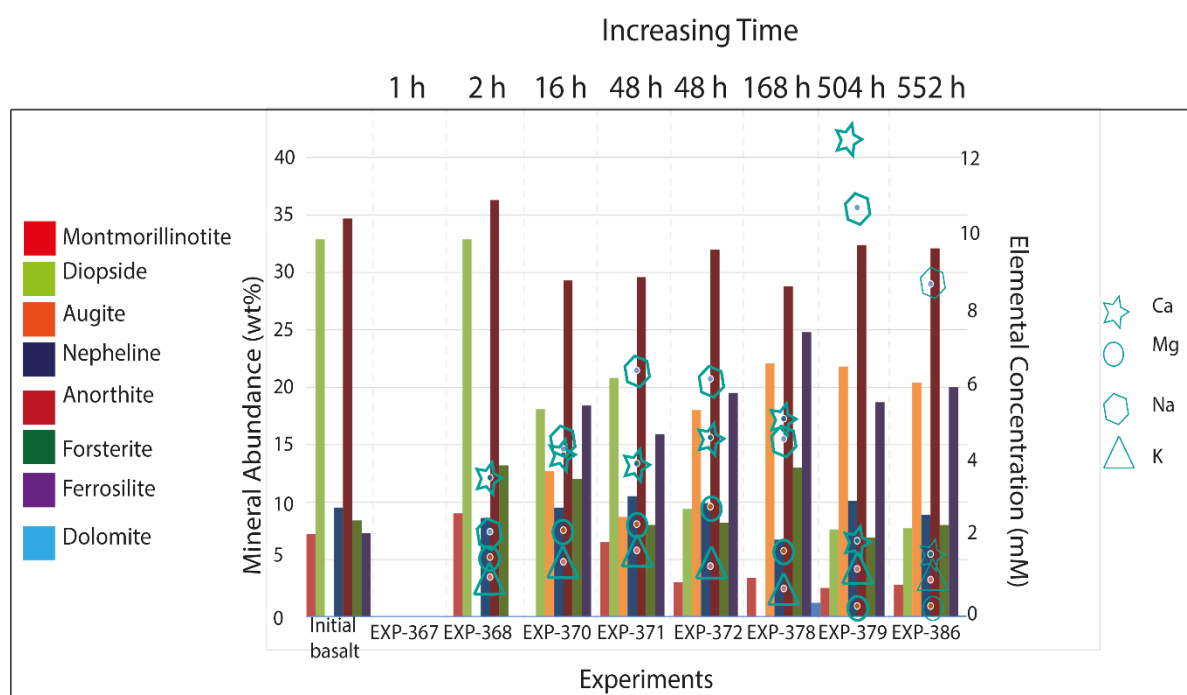


Fig. 6 - Image shows XRD analysis results of basalt sample and post-experiment water analysis, graph showing initial sample and post-experiment samples (x axis), mineral abundances (wt. %) (left y axis), and element concentration of post-experiment analysis (right y axis). Legend on left side shows in circle major elements, legend on right side shows the dissolved Ca, Mg, Na, and K concentrations measured by ICP-OES and expressed in mM. A colored version of the picture is available online.

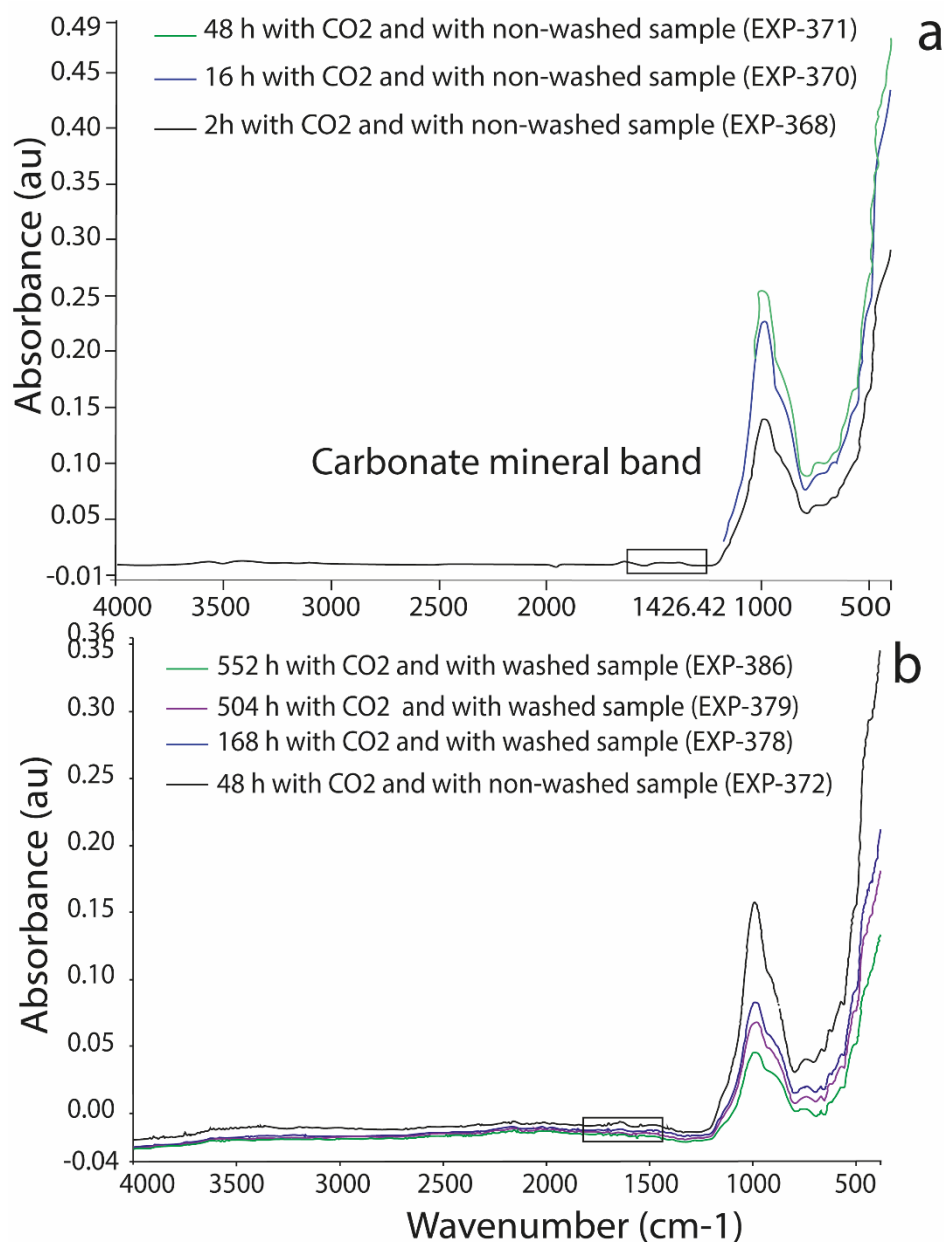


Fig. 7 - Image shows post-experiment FTIR spectra of reacted basalt samples following ageing experiments (a) short-duration experiments EXP-368-371 (2-48 h) using non-washed samples and (b) longer-duration experiments EXP-372-386 (48h - 552 h). The boxed region highlights the carbonate absorption band at approximately 1426 cm⁻¹. A colored version of the picture is available online.

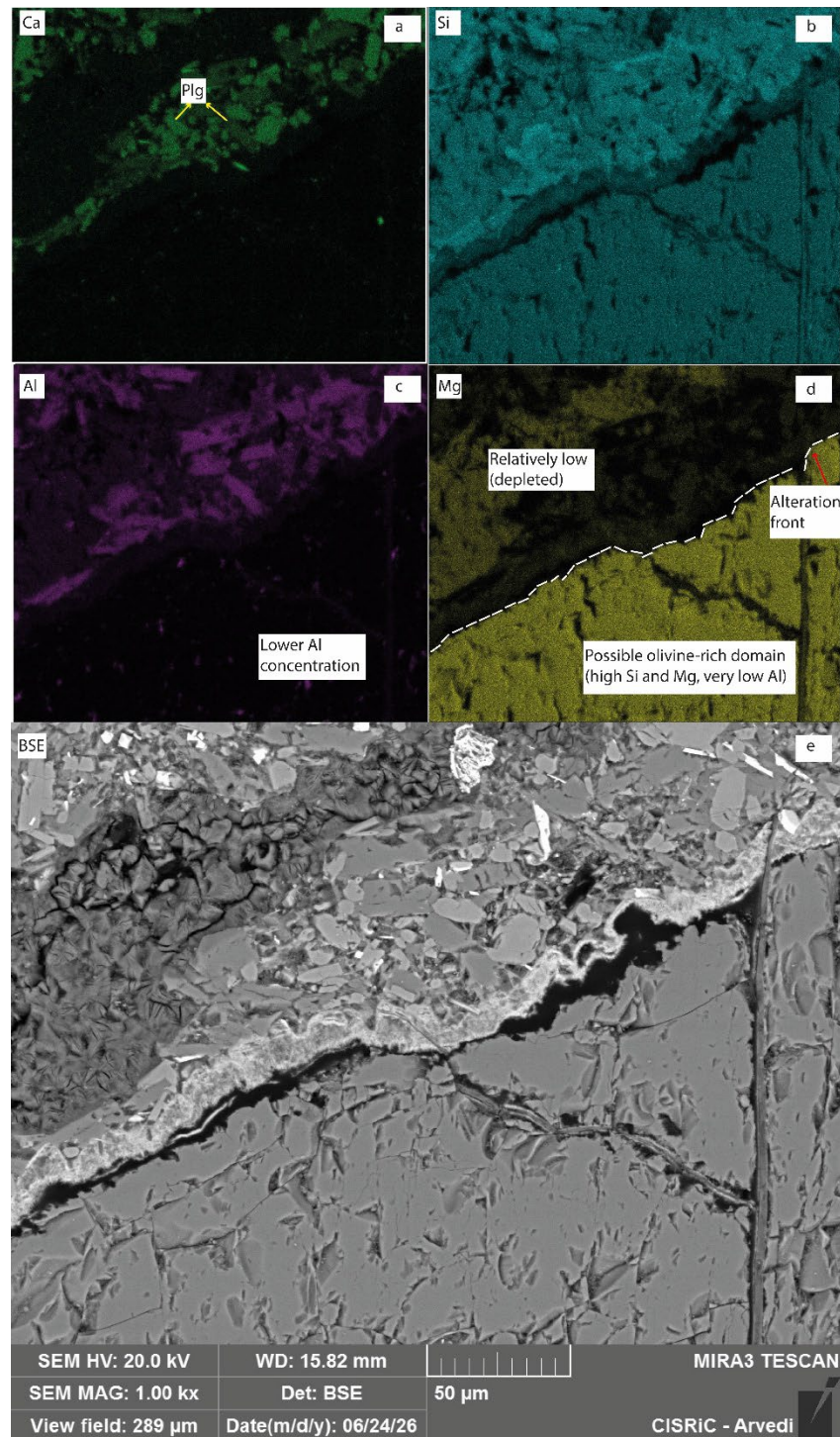


Fig. 8 - SEM-EDS elemental maps showing the spatial distribution of Ca (a), Si (b), Al (c), and Mg (d), together with the corresponding BSE image (e) of the starting basalt (DIS14-506). Ca enrichment highlights plagioclase microliths, whereas the elemental maps reveal compositional heterogeneity between the upper and lower domains. The lower domain is characterized by relatively higher Mg and lower Al contents, while the upper domain exhibits Mg depletion, Al enrichment, and a more heterogeneous texture. The dashed line indicates the transition between the two chemically distinct domains, interpreted as an alteration front. A colored version of the picture is available online.

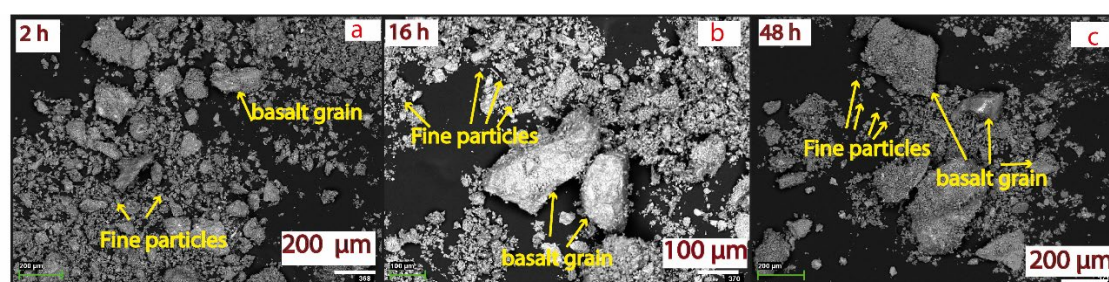


Fig. 9 - SEM images of post-experiment unwashed samples (a) EXP-368 (2 h); (b) EXP-370 (16 h); (c) EXP-371 (48 h). A colored version of the picture is available online.

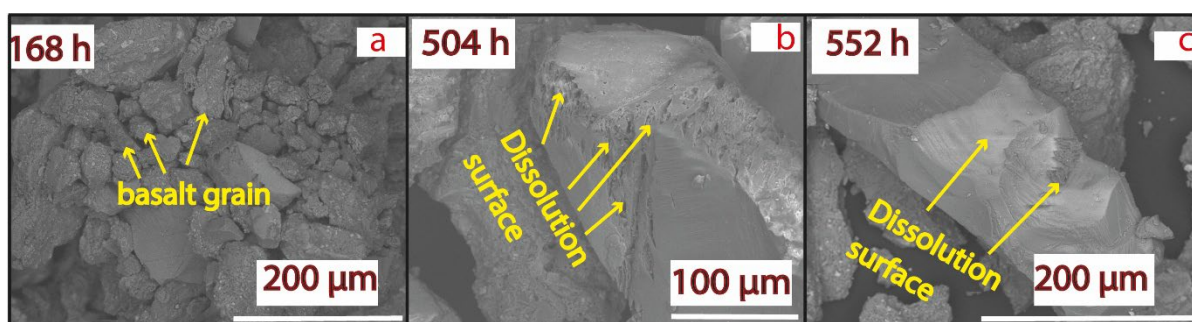


Fig. 10 - SEM images of washed post-experiment samples (a) EXP-378 (168 h); (b) EXP-379 (504 h); (c) EXP-386 (552 h). A colored version of the picture is available online.

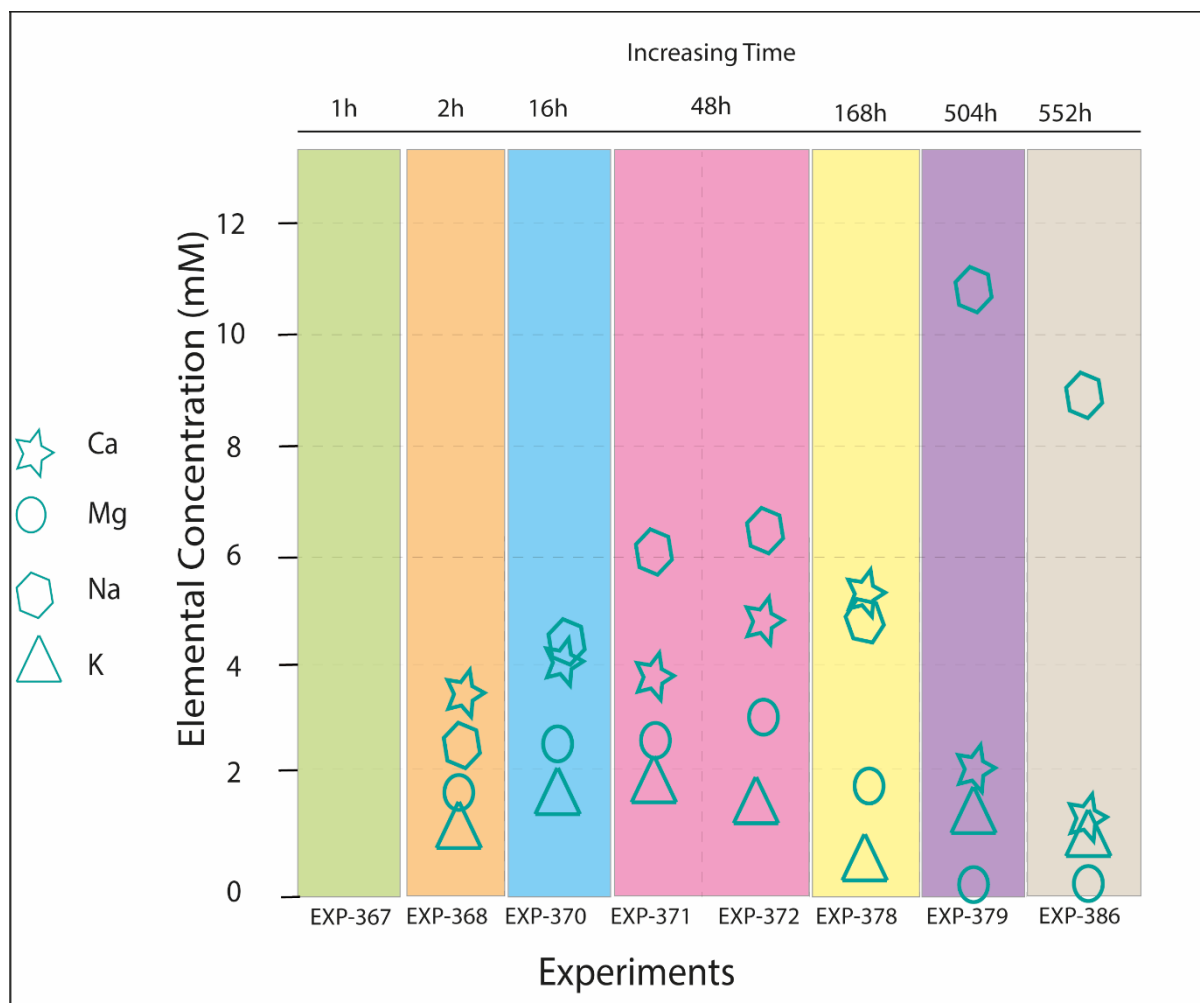


Fig. 11 – Image shows evolution of post-experiment liquid analysis after each ageing experiment measured by ICP-OES. X- axis indicates carried out experiments and arranged according to increasing reaction time from 1 to 552 h and y-axis indicates elemental concentrations in mM. Colored backgrounds distinguish the individual experiments. Different symbols represent each analyzed major elements. A colored version of the picture is available online.

Table 1 - Experimental conditions and dissolved element concentrations measured by ICP-OES after the ageing experiments.

<i>Exp</i>	367	368	370	371	372	378	379	386
<i>Total Duration (h)</i>	1	2	16	48	48	168 (72)	504 (168)	552 (502)
Liquid results :								
mM								
<i>Ca</i>	0.05	3.64	4.24	4.02	4.69	5.19	1.99	1.65
<i>Mg</i>	0.07	1.56	2.26	2.43	2.88	1.73	0.29	0.29
<i>Na</i>	0.03	2.22	4.39	6.44	6.22	4.65	1.07	8.7
<i>K</i>	0.01	1.04	1.43	1.74	1.33	0.74	1.25	0.97
µM								
<i>Al</i>	7.78	3.07	5.48	5	218	2.33	13.71	9.64
<i>Ti</i>	0.23	10.17	10.71	9.04	0.96	<0.21	<0.21	<0.21
<i>V</i>	<0.08	0.1	0.27	0.39	0.23	0.14	<0.08	<0.08
<i>Cr</i>	<0.96	<0.96	<0.96	<0.96	3.94	<0.96	<0.96	<0.96
<i>Mn</i>	6.73	32.43	25.9	27.37	43.68	28.03	15.83	14.2
<i>Fe</i>	14.2	3.49	6.8	7.48	73.42	21.92	143.25	69.84
<i>Ni</i>	121.26	15.47	9.76	13.83	30.67	594.82	102.23	66.45
<i>Cu</i>	4.48	<0.31	<0.31	<0.31	0.6	<0.31	<0.31	<0.31

Values in parentheses indicate the duration of the low-pressure stage following depressurization.
Major elements are reported in mM/L, whereas trace elements are reported in µmol/L.

Table 2– Bulk characterization (wt.%) analysis of the basalt pre- and post- experiment (EXP -378 (168 h), EXP-379 (504 h) and EXP -386 (552 h)) by XRF analysis

	DIS14-506	EXP378	EXP379	EXP386
SiO ₂	44.54	44.66	45.19	44.89
TiO ₂	2.68	2.53	2.59	2.60
Al ₂ O ₃	14.98	14.21	14.48	13.80
FeO _(T)	8.38	8.38	8.36	8.78
MnO	0.16	0.17	0.17	0.17
MgO	10.13	12.28	11.47	12.27
CaO	10.03	10.02	10.07	9.95
Na ₂ O	3.38	4.02	4.04	3.69
K ₂ O	1.49	1.17	1.09	1.10
P ₂ O ₅	0.70	0.67	0.69	0.66
L.O.I.	2.81	1.89	1.86	2.09
Total	99.28	100.00	100.01	100.00

Table 3 - Summary of selected batch experiments investigating CO₂ mineralization in basalt under different experimental conditions.

References	T (°C)	P (bar)	Time	Carbonate precipitation
Wang et al., 2024	100	50	360 h	No
Clark et al., 2019	50	80	180 days	No
Galeczka et al., 2013	22	80	50-100 h	No
Brady and Gislason, 1997	25, 37 and 50	3.2 x 10 ⁻⁴ , 5.08 x 10 ⁻³	one month	No
Menefee et al., 2018	100 and 150	100	10 and 12 and 12 days	Yes
Kumar et al., 2017	100	5, 10	50, 60, 70, 80, 100 h	Yes
Berndsen et al., 2024	105, 260	8, 170	30 days	No
Hellevang et al., 2017	80, 100, 150		21 to 52 days	Yes at 150 °C
Hovellmann et al., 2012	100-150-200	14.6- 19.4	7, 21 and 42 days	Yes
Kikuchi et al., 2023	200, 230, and 300	85	5 and 10 days	Yes
Rosenbauer et al., 2012	50-200	300	Until 2064 h	Yes
Giammar et al., 2005	30 and 95	1-103	Until 630	Yes

The table highlights the wide range of conditions under which carbonate mineralization has been investigated and provides a comparison with the reaction durations considered in the present study.