

Modelling Migration of CO₂ in Fractures Basalt Reservoirs – Preliminary Testing of a Novel Multi-physics Reactive-Flow Model

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The geological storage of CO₂ is a promising means to help reduce the emissions of greenhouse gases into the atmosphere during the energy transition stages (e.g., Raza *et al.*, 2022). Carbon Capture and Storage (CCS) is one of the essential technologies required to achieve global temperature rise limitations (IPCC, 2022) by contributing to the mitigation of carbon emissions from energy and industrial plants during the transition and is furthermore expected to support direct CO₂ removal from the atmosphere on a longer timescale (Snaebjörnsdóttir *et al.*, 2020).

Storage of CO₂ in geological formations depends on a combination of physical and chemical mechanisms such as physical trapping below caprocks or trapping by dissolution in groundwater. The most effective storage mechanism is the permanent mineralisation of CO₂ by conversion into carbonate minerals (Benson *et al.*, 2005). The relative trapping mechanisms contributions change through time after CO₂ injection (Figure 1). Basalts have an excellent storage potential because they are widespread around the world and because the mineralisation of injected CO₂ is completed within a decade or less, reducing leakage risks to a minimum (Oelkers *et al.*, 2023).

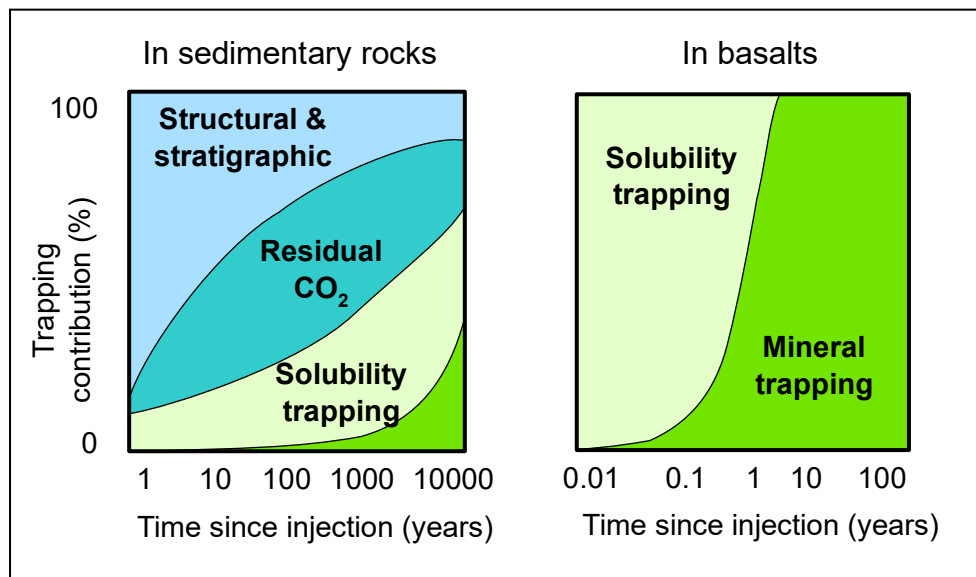


Figure 1: Relative contributions through time of different trapping mechanisms in sedimentary rocks (left) and basalts (right) (modified after Benson *et al.*, 2005).

Monitoring of potential surface emissions of CO₂ escaped from geological reservoirs is an integral part of industrial CO₂ storage activities. It is, for instance, required from operators in the EU legislation (Directive 2009/31/EC). However, monitoring of CO₂ plumes in the subsurface relies essentially on very cost-intensive 4D seismic imaging and/or drilling boreholes. Furthermore, appropriate surface monitoring methods and strategies are still the object of intense research due to the complexity of the problem (e.g., Blackford *et al.*, 2021) and Measurement, Monitoring and Verification (MMV) activities will be costly too.

At IGI, we have recently initiated an R&D project to develop a novel modelling protocol to model the migration of CO₂ between potentially failing caprocks, or leakage due to failures in the infrastructure (e.g. borehole casing), and the surface. Our aim is to bridge the gap between reservoir and surface monitoring, thus contributing to the optimisation of caprock integrity monitoring and of MMV deployment, driving significant costs reductions. Our R&D project involves the adaptation of a state-of-the-art multi-physics reactive flow numerical model which was produced for the investigation of the dynamic behaviour of gas hydrates in marine sediments (Gupta *et al.*, 2020, 2023; Schmidt *et al.*, 2022).

Here, we present the preliminary results of the first tests performed to assess the ability of the early numerical model to handle coupling of geochemical and hydrodynamic processes (Nuzzo *et al.*, 2024). We built a box-model for a simple basalt carbonation scenario, which has been selected because it involves notably complex chemical reaction networks that will support the future development of highly comprehensive geochemical models.

Carbonation occurs through the interaction of water-dissolved CO₂ and formation minerals, which involves mineral dissolution followed by carbonate precipitation (Figure 2). Strongly acidic CO₂-rich fluids (pH ~ 3 to 5) promote the dissolution of silicates through the consumption of protons. The disaggregation of silicate minerals releases cations, which bind with carbonate anions as pH values rise due to the buffering effect of silicates dissolution (e.g., Oelkers and Gislason, 2001; Heřmanská *et al.*, 2021; Snaebjörnsdóttir *et al.*, 2020).

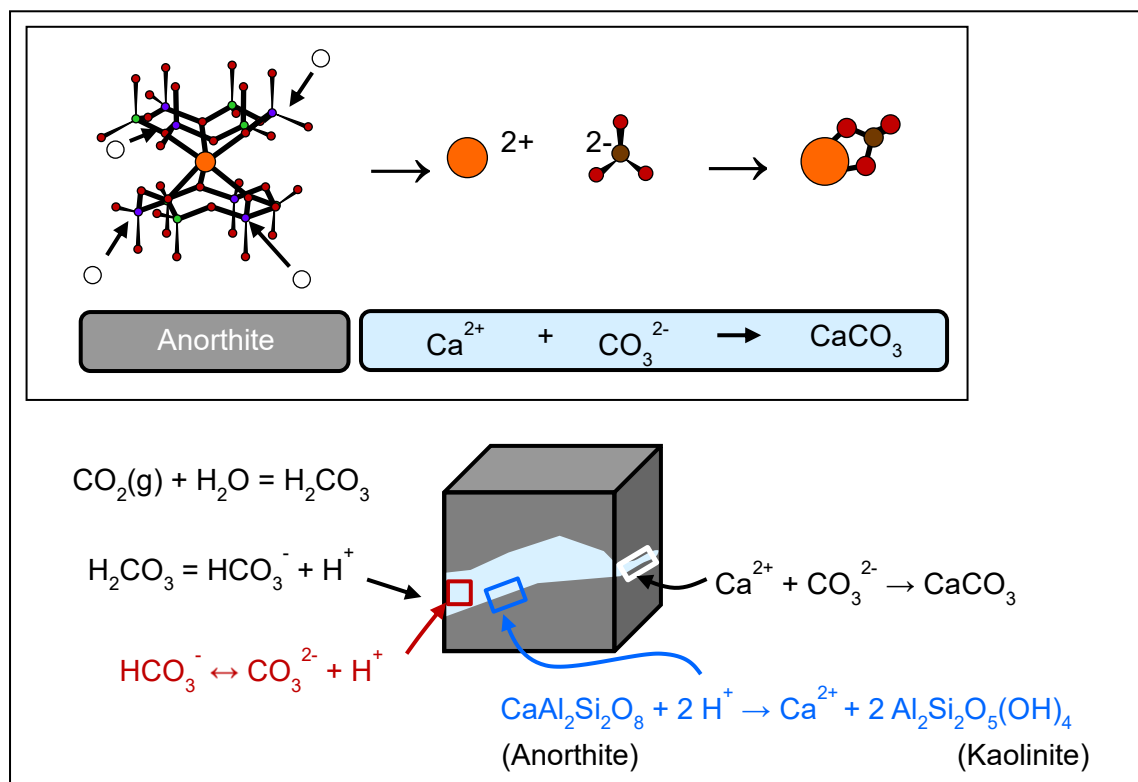


Figure 2: Schematic illustration of the related processes of dissolution of silicate minerals (here anorthite) by low pH in CO₂-rich water and precipitation of carbonate minerals.

Whilst fractures in lava flows provide reactive surfaces for CO₂ mineralisation, they also represent high permeability pathways for potential escape of CO₂ until secondary carbonates “self-seal” the fractured reservoir. Our experiment aims at simulating changes in porosity

accompanying mineral dissolution-precipitation reactions and the consequent fluid flow field modifications. Through this, we evaluate how the numerical model performs with respect to coupling of geochemical and hydraulic parameters. In the 2D simplified model, the lava flow is represented by two colonnade layers separated by an entablature interval and liquid CO₂ is injected into the lower colonnade (Figure 3).

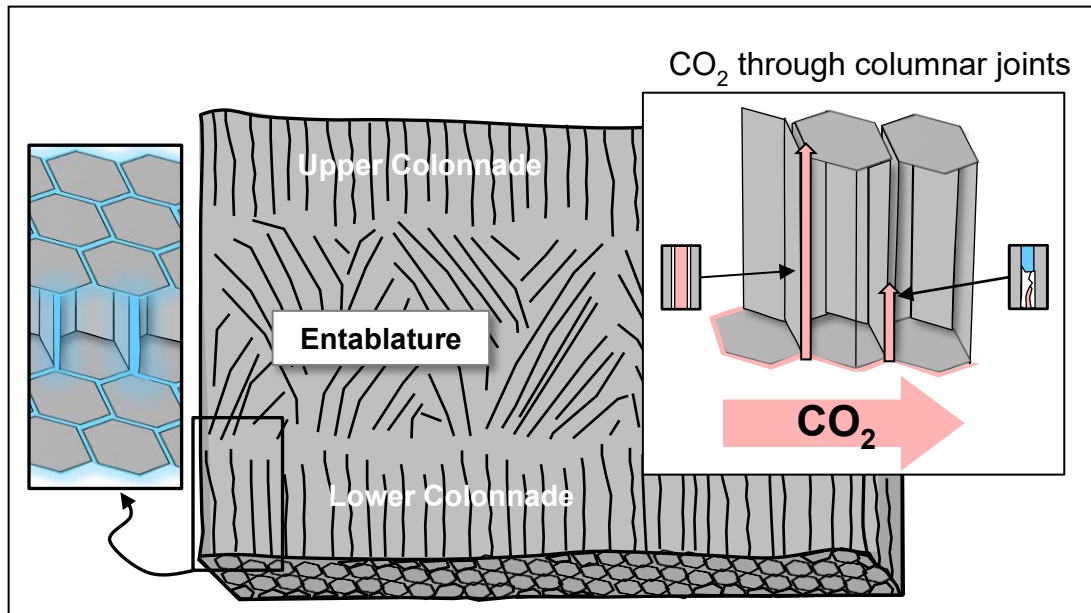
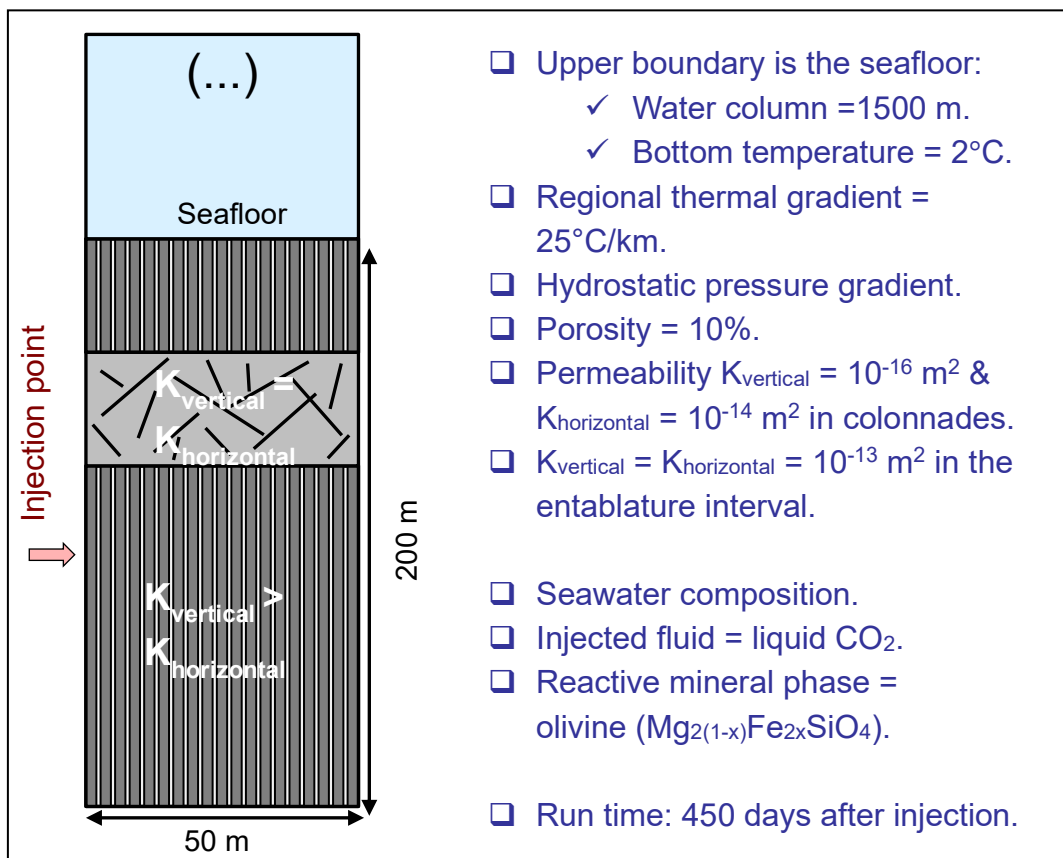


Figure 3: Schematic depiction of CO₂ fluids migration in fractures between basaltic columns.

The reactive phase is represented in this model by olivine, with this simple (non-realistic) framework enabling a better understanding of how the numerical model functions. Main model parameters are indicated below.



- ❑ Upper boundary is the seafloor:
 - ✓ Water column = 1500 m.
 - ✓ Bottom temperature = 2°C.
- ❑ Regional thermal gradient = 25°C/km.
- ❑ Hydrostatic pressure gradient.
- ❑ Porosity = 10%.
- ❑ Permeability $K_{\text{vertical}} = 10^{-16} \text{ m}^2$ & $K_{\text{horizontal}} = 10^{-14} \text{ m}^2$ in colonnades.
- ❑ $K_{\text{vertical}} = K_{\text{horizontal}} = 10^{-13} \text{ m}^2$ in the entablature interval.
- ❑ Seawater composition.
- ❑ Injected fluid = liquid CO₂.
- ❑ Reactive mineral phase = olivine ($\text{Mg}_{2(1-x)}\text{Fe}_{2x}\text{SiO}_4$).
- ❑ Run time: 450 days after injection.

In the test presented here, simulations have been run with dissolution reaction rate of olivine varying by two orders of magnitude between the “low reactivity/dissolution rate” and the “high reactivity/dissolution rate” endmembers. The influence of the silicate mineral reactivity on the evolution of the plume and on porosity through time is very significant as shown on Figure 4.

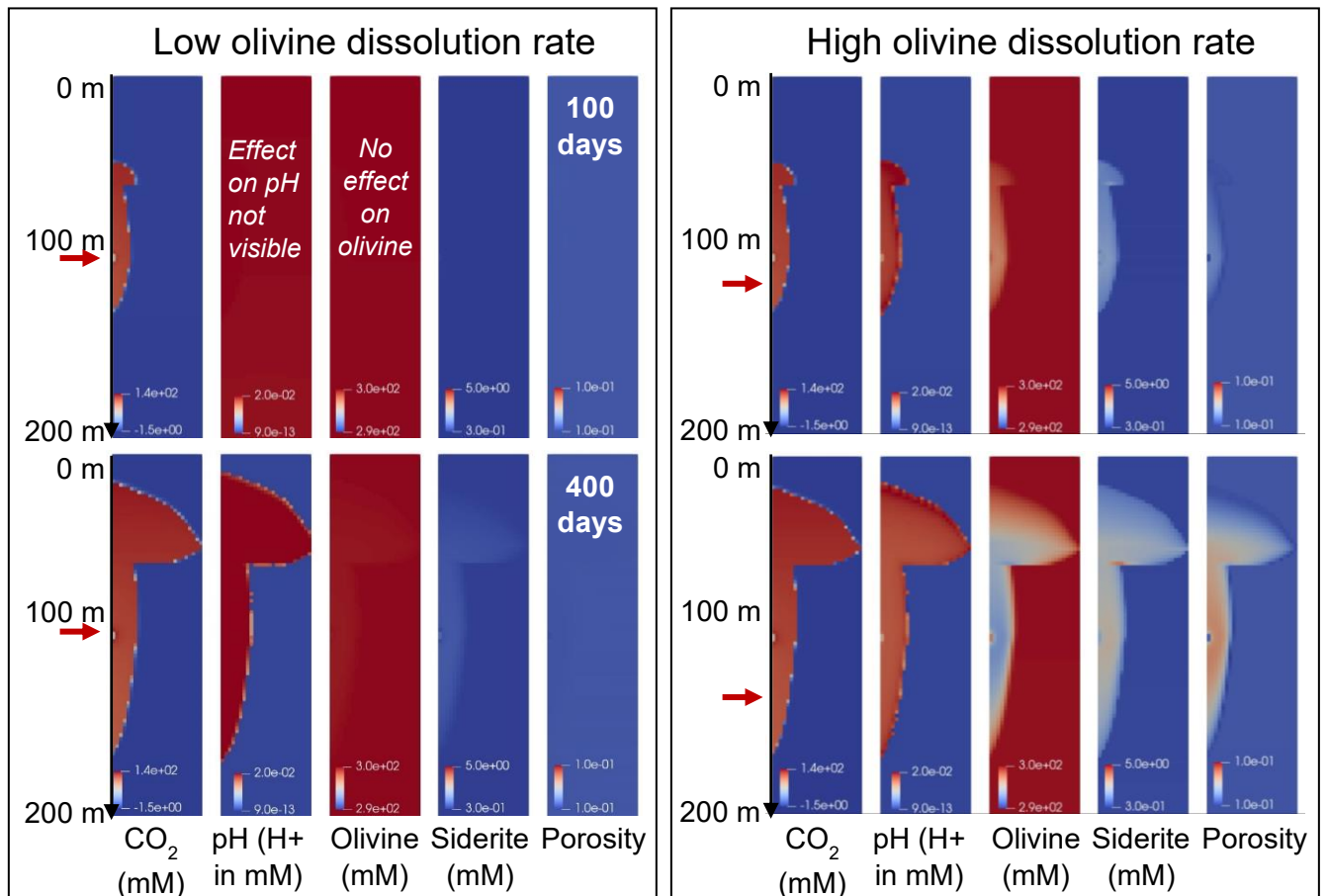


Figure 4: Evolution of selected parameters as the injected CO₂ plume develops in the subsurface 100 days (top) and 400 days (bottom) after the start of injection. The results show the simulation with low and with high mineral reactivity (i.e., olivine dissolution/precipitation rates) on the left and on the right, respectively. The propagation of the plume below 100 m is due to the set-up of the numerical model and is not meaningful.

The spatial evolution of the plume (CO₂ concentration) is strongly correlated with permeability contrasts between layers, with the main flow direction changing from vertical in the lower colonnade ($K_{\text{vertical}} > K_{\text{horizontal}}$) to diagonal in the entablature ($K_{\text{vertical}} = K_{\text{horizontal}}$). Different authors have reported the permeability of entablatures to be lower or, on the contrary, higher than that of colonnade intervals (e.g., McGrail *et al.*, 2009). In this study, we have assumed a higher permeability entablature layer given that only the permeability contrast between layers is of relevance for the development of the numerical model. Increased CO₂ concentration drives the sharp pH decrease in the plume area.

Decreasing olivine concentrations in the CO₂ plume results promotes siderite concentration (FeCO₃) as the result of the reaction of iron (Fe²⁺) released from olivine (Mg_{2(x-1)}Fe_{2x}SiO₄) with carbonate ions (CO₃²⁻). The magnitude of the change in olivine and siderite concentrations is

strongly affected by the increase in olivine dissolution rates (“reactivity”). The pH buffering effect of olivine dissolution is obvious in the significantly lower pH of the plume in the low olivine reactivity test. While the absolute porosity change is extremely low (0.099 to 0.1004), olivine dissolution is shown to drive porosity increases.

The box model presented here shows that the numerical code resolves the coupling of hydraulic and chemical reactivity processes associated with evolving porosities in the plume. Furthermore, it demonstrates that it can robustly handle phase transitions consistently with pore pressure and temperature changes.

Encouraged by these results, we will proceed with the incremental complexification of the numerical model. Ultimately, we will develop a novel multiphysics reactive flow CO₂ migration modelling protocol with a unique capacity to apprehend complex non-linearly related processes that condition the evolution of the CO₂ plume in the subsurface at basin-scale (CO2MIG project <https://igilt.com/news/geological-storage-of-co2-carbon-capture-and-storage-ccs>).

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