Dissolution experiments on a polyphase basalt surface under conditions relevant to offshore CO_2 storage

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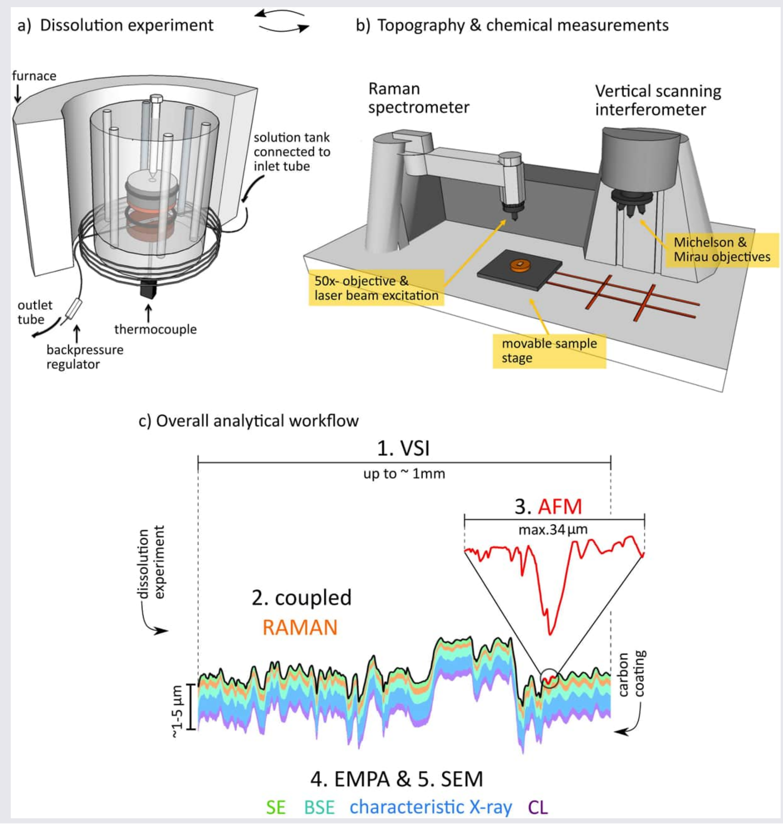
MOTIVATION

Basalt dissolution rates are critical for carbonate mineralization. Many laboratory experiments have been conducted on a multitude of rock forming minerals to investigate the dissolution kinetics and mechanisms under various experimental conditions (Heřmanská et al., 2022a). However, dissolution rate quantification based on fluid composition measurements, which provide mean dissolution rates, are limited in their ability to provide information about dissolution rate variabilities due to material heterogeneities, e.g., chemical composition, crystallographic properties, and texture (Blum and Stillings, 1995). We measure dissolution rates based on spatially resolved changes in surface topography and apply complementary techniques to obtain information on chemistry, structure and texture of the dissolving surface.

METHODS



Experimental set up and workflow (Lange et al., 2021)

> polished, micro crystalline basalt surface reacted at 70°C for ca. 6 weeks with CO_2-charged ultra pure water

> measurements of surface topography before and after dissolution experiment

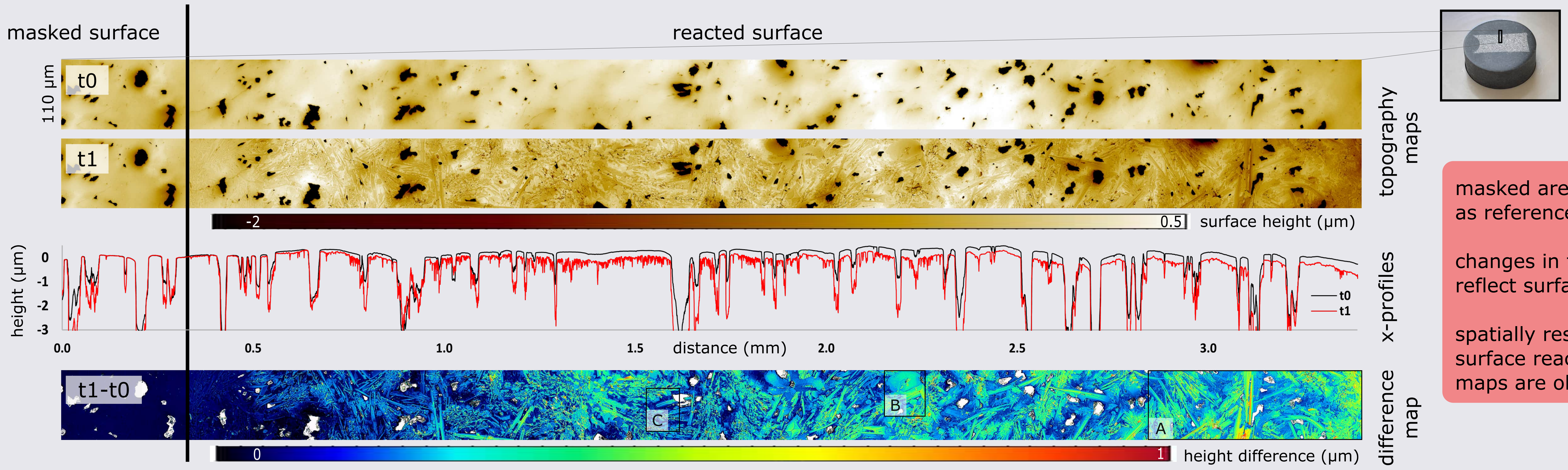
> part of the surface is masked, thus, not in contact with fluid, and serves as reference height

> dissolution rates are calculated based on absolute height changes (Δh) divided by the experimental duration (Δt) and the molar volume (Vm) of the material

> Raman spectroscopy and electron beam techniques provide information on chemical composition, structure, and texture

RESULTS

1 SURFACE TOPOGRAPHY MEASUREMENTS



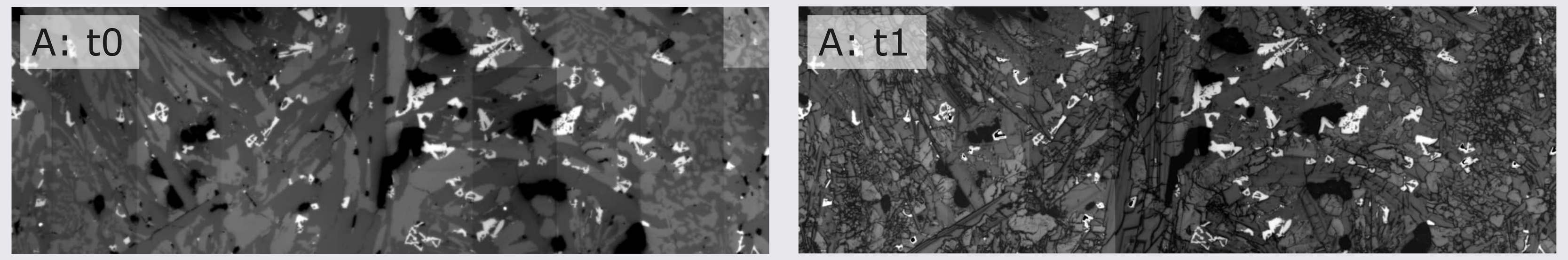
masked area serves as reference height

changes in topography reflect surface reactivity

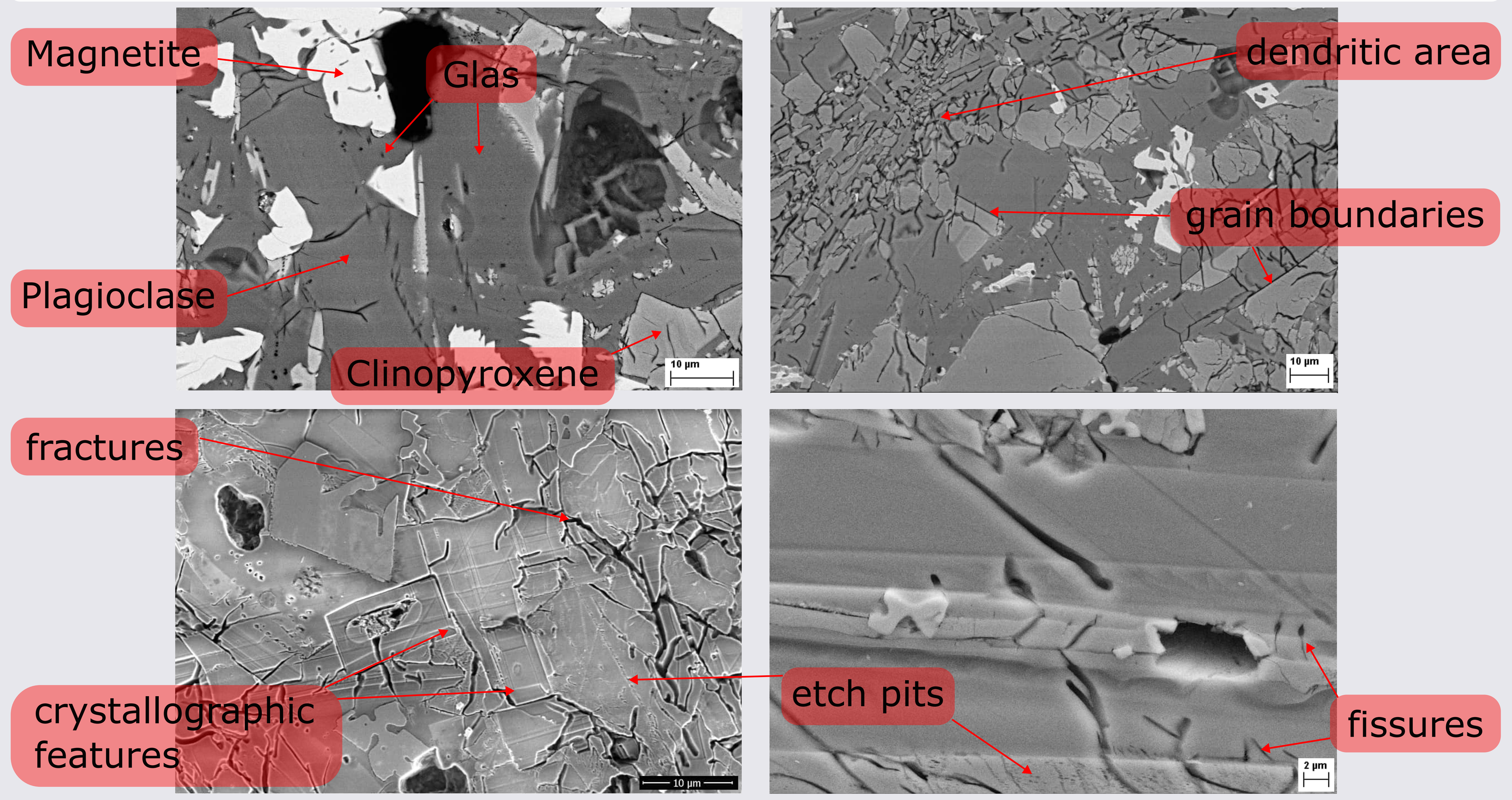
spatially resolved surface reactivity maps are obtained

t0: initial, polished surface topography, t1: topography after dissolution experiment, x-profiles of surface topographies t0 and t1, t1-t0: absolute height differences

2 MINERAL PHASES & TEXTURE

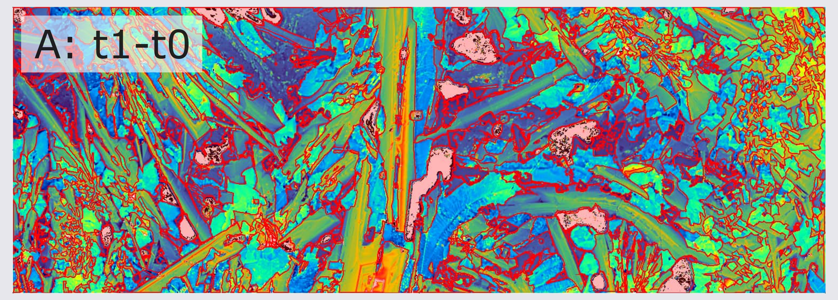


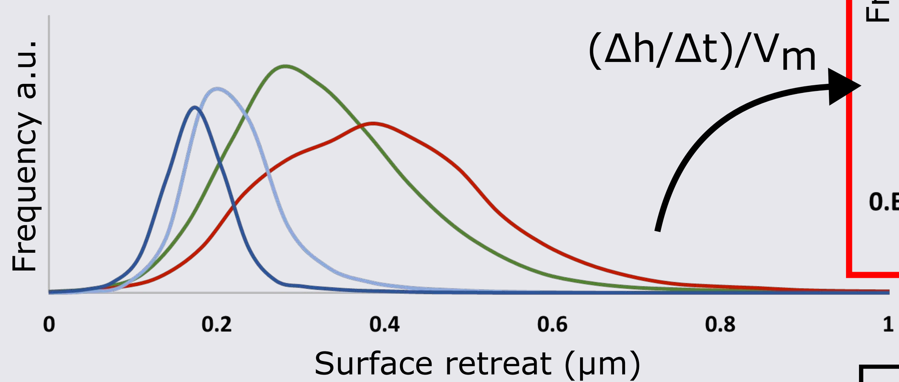
Light reflection images before and after the dissolution experiment showing a polymineralic surface with dissolution induced surface features.



BSE (upper) and SE (lower) images showing multiple dissolution induced features such as etch pits and fractures on the surfaces and along grain boundaries.

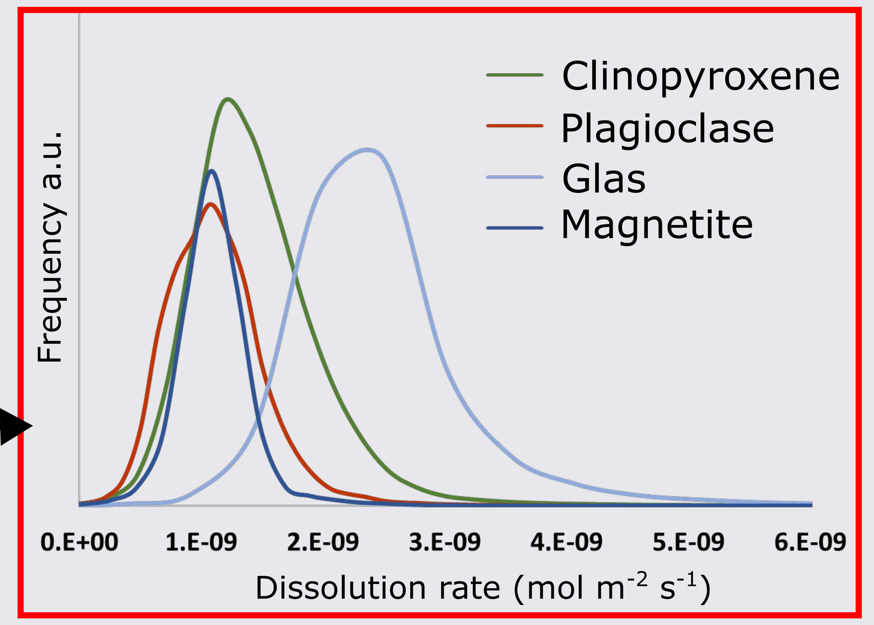
3 MINERAL DISSOLUTION RATES





Surface retreat and dissolution rate histograms were calculated based on mineral phase segmentation of the difference map.

(Δh/Δt)/Vm

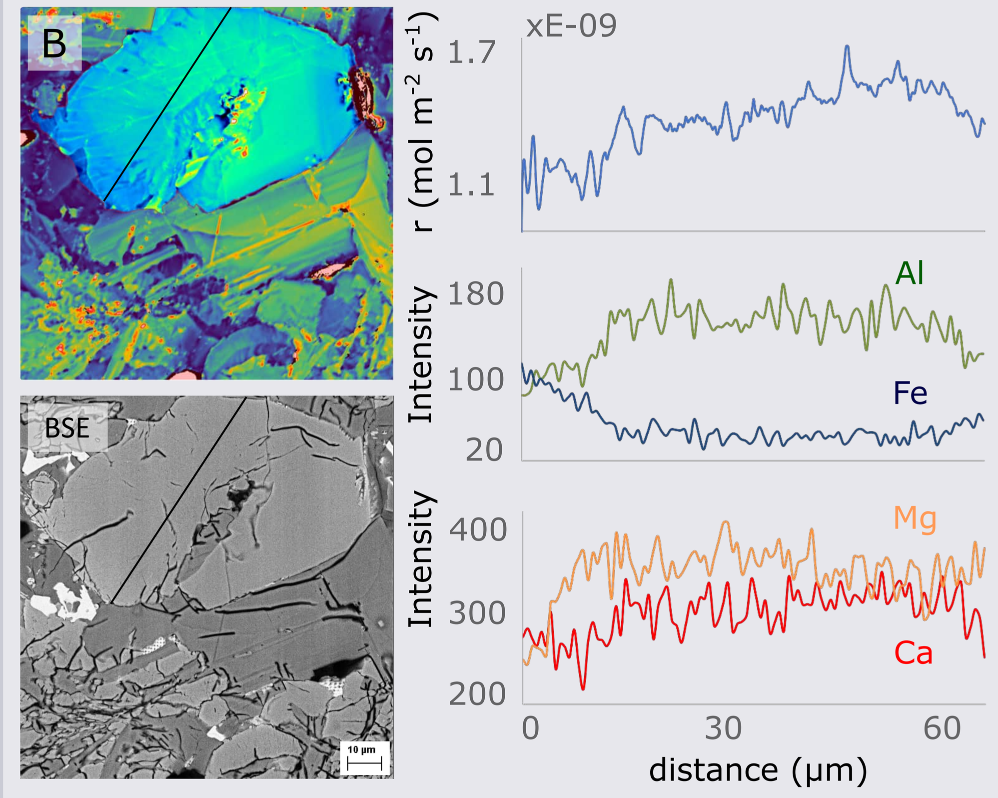


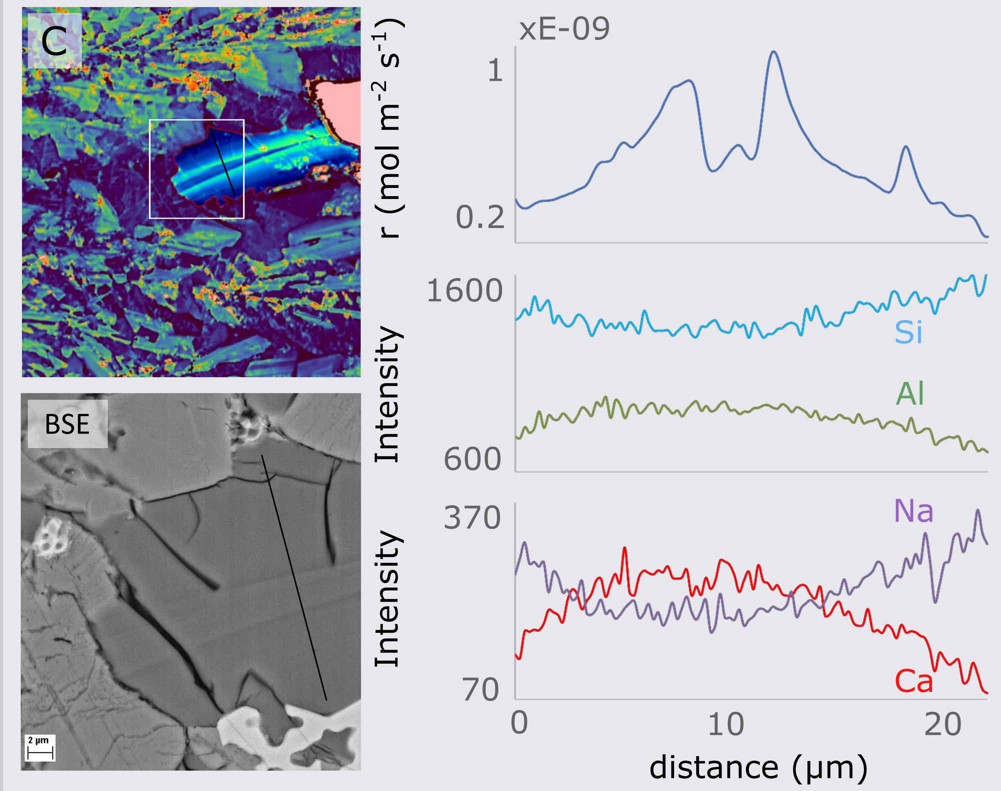
	mode rate (mol m^{-2} s^{-1})
Magnetite	1.08E-09
Plagioclase	1.08E-09
Clinopyroxene	1.17E-09
Glas	2.49E-09

The order of the histograms changes when the retreat values are divided by the respective molar volumes.

The seperate phases show differences in dissolution rates and their distributions.

4 CRYSTAL HETEROGENEITY





Dissolution rate and EDX profiles of a pyroxene (upper) and a plagioclase (lower) crystal. The rates vary with chemical composition.

CONCLUSIONS

> spatially resolved dissolution rates of rock forming minerals are obtained within one experiment under identical conditions

> mineral phases show differences in dissolution rates and rate distributions

> dissolution rate variabilities within single crystals are resolved

> data show strong influence of, e.g., grain boundaries, chemical composition, and twinning

References: **[1]** Heřmanská , M., Voigt, M.J., Marieni, C., Declercq, J., Oelkers, E.H., 2022a. A comprehensive and internally consistent mineral dissolution rate database: Part I: Primary silicate minerals and glasses. *Chemical Geology* 597, 120807. **[2]** Blum, A.E. and Stillings, L.L., 1995. Feldspar Dissolution Kinetics. In: Chemical Weathering Rates of Silicate Minerals. *Reviews in Mineralogy*, Vol. 31. Ed. by AF White and SL Brantley. Berlin, Boston. Chap. 7, pp. 291–352. **[3]** Lange, I., Toro, M., Arvidson, R.S., Kurganskaya, I., Luttge, A., 2021. The role of crystal heterogeneity in alkali feldspar dissolution kinetics. *Geochimica et Cosmochimica Acta* 309, 329-351.