Electronic Supplementary Information

Emerging investigator series: Kinetics of Diopside Reactivity for Carbon Mineralization in Mafic-Ultramafic Rocks

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Table S1. Surface Me-O (Me=Ca,Mg) coordination numbers (CN) on the (110) surface of diopside.

Case	Surface Energy γ (J/m^2)
(I)	1.1199
(II)	0.6344
(III)	1.0814
(IV)	1.7767

Table S2. Surface Me-O (Me=Ca,Mg) coordination numbers (CN) on the (110) surface of diopside.

	Case (II)		Case (III)		
Surface ion	CN (unrelaxed)	CN (relaxed)	CN (unrelaxed)	CN (relaxed)	CN (bulk)
Ca	5	6	7	8	8
Mg	5	6	4	6	6

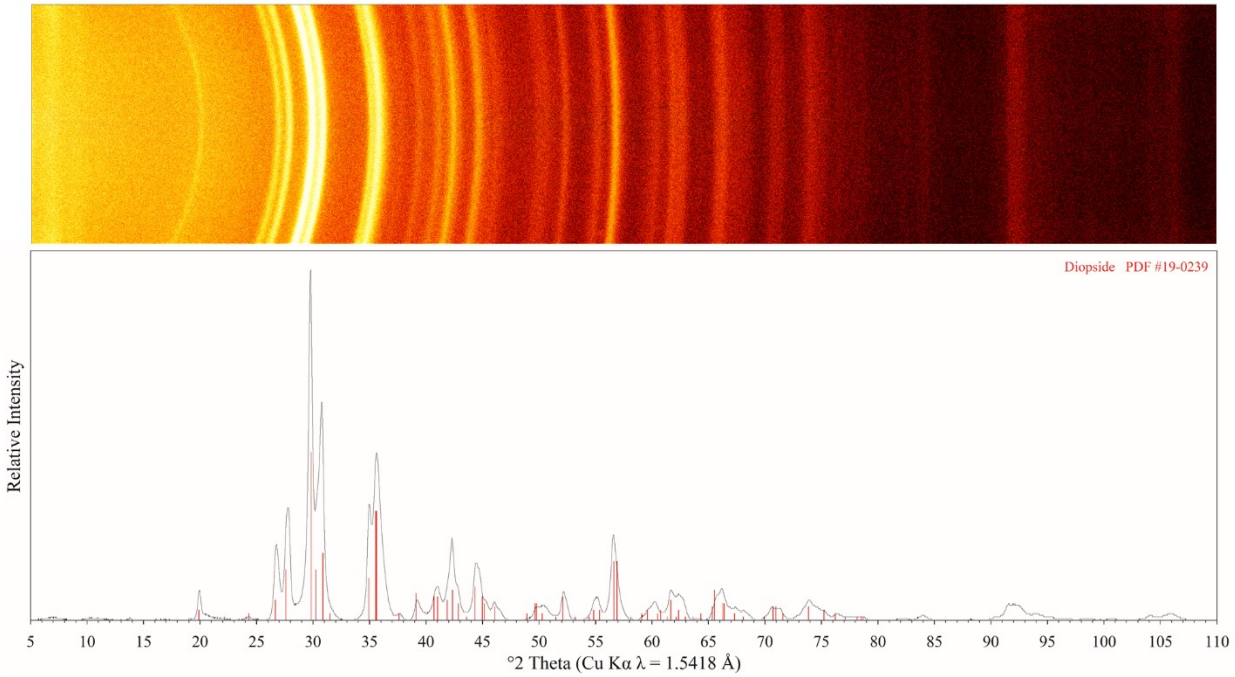

Figure S1. Reference XRD pattern for Tajuelo Rodriguez et al.[1] diopside sample used in **EXP I-IX**. XRD scans of unreacted diopside collected using a Bruker D8 Discover TXS-HE A25, equipped with a rotating Cu anode (Kα λ = 1.5418 Å), 0.3x3 mm cassette tungsten filament, Atlas goniometer, and a UMC 1516 motorized stage. A powder sample was mounted on the xyzϕχ stage and positioned using a laser-video alignment system. The power settings of the generator were 45 kV and 120 mA and the source-sample distance is fixed at 425 mm. The EIGER2 R 500K detector was positioned at a 206.8 mm sample-detector distance. The upper 2D detector image was collected with the EIGER in max γ orientation via a continuous coupled 2θ/θ scan from 5-110 °2θ with a 0.02 step size and 0.25 time/step while the stage rotated (ϕ) at 360 °/min. The ~2 mm point source was generated using a Montel mirror optic and a 2 mm collimator. The lower diffractogram was collected with the detector in 1D max 2θ mode orientation with a mounted 78x25 mm panoramic axial Soller (2.5°). Collection of individual XRD tracings from 5-110 °2θ in coupled 2θ/θ mode with 5251 steps at 3.5 seconds per step, X-Y grid scanning (4 mm amplitude at 2 mm/s and 2 mm amplitude at 4 mm/s, respectively), and continuous stage rotation (ϕ) at 360 °/min. On the source side, the TXS-HE line beam passed through a focusing Goebel mirror, a 0.6 mm divergence slit, a 2.5° axial Soller, and a 22 mm-long scatter guard.

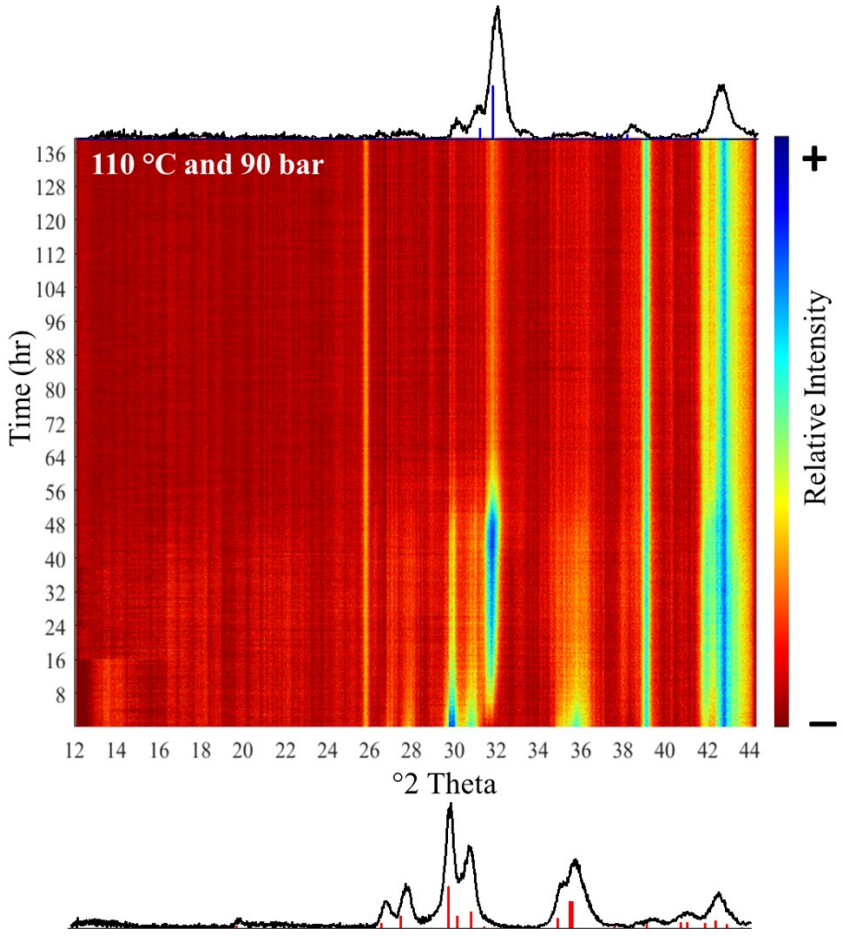

Figure S2. Full 2D waterfall plot for **EXP I** (sample fell from holder at ~56 hours).

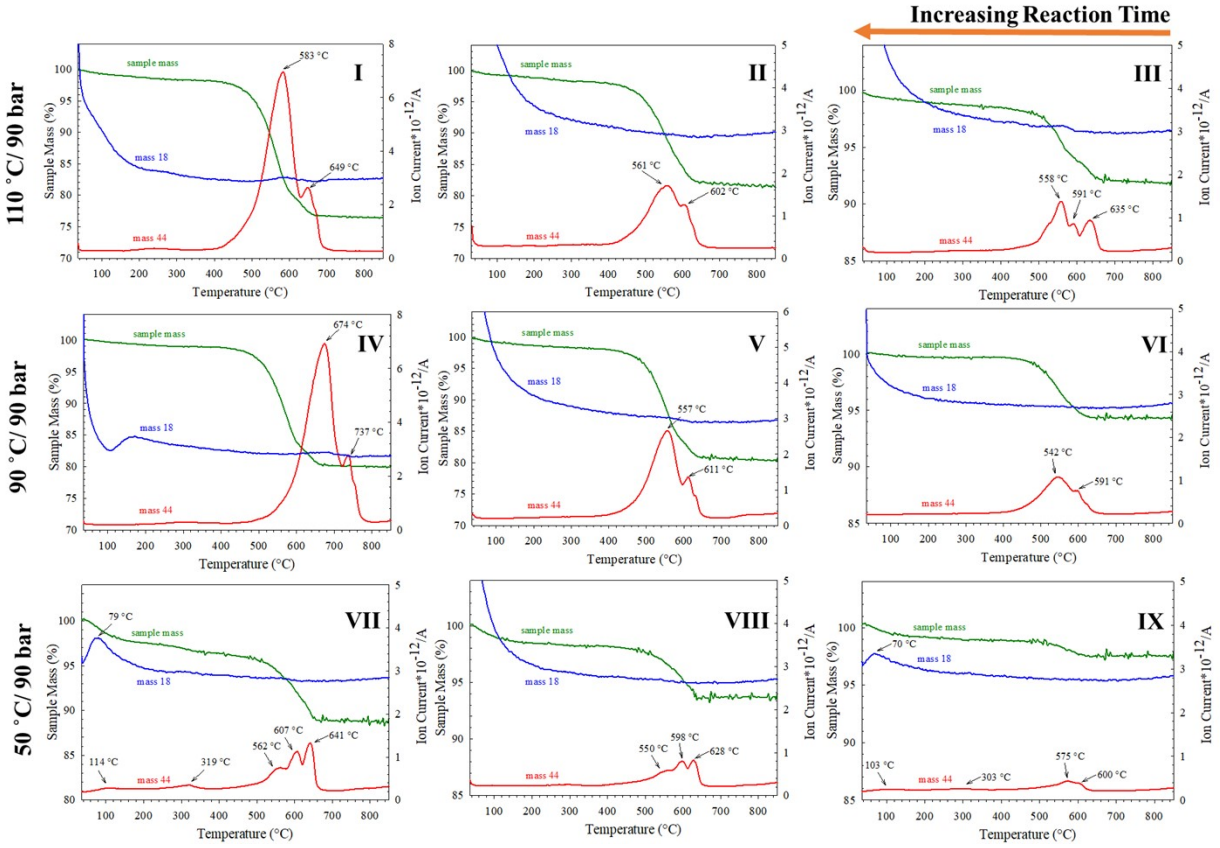

Figure S3. Thermogravimetric mass spectrometry measurements for post-reaction samples from **EXP I-IX** (m/z=18 & 44).

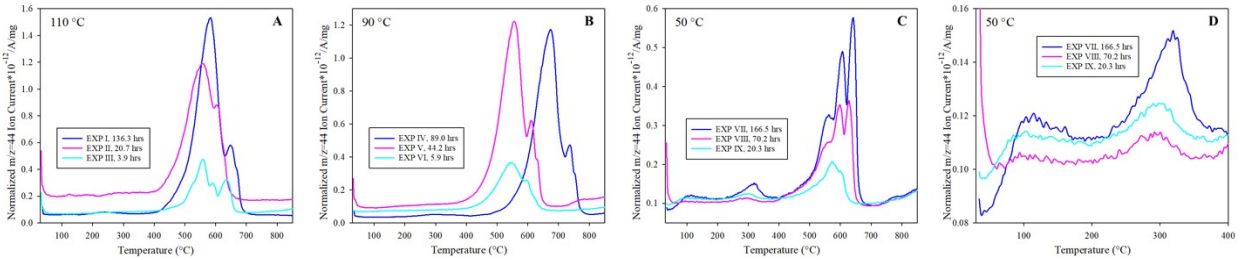

Figure S4. Mass-normalized mass spectrometry (m/z=44) results for (A) **EXP I-III**, (B) **EXP IV-VI**, (C) **EXP VII-IX**, and (D) the 35-400 °C range for **EXP VII-IX**.

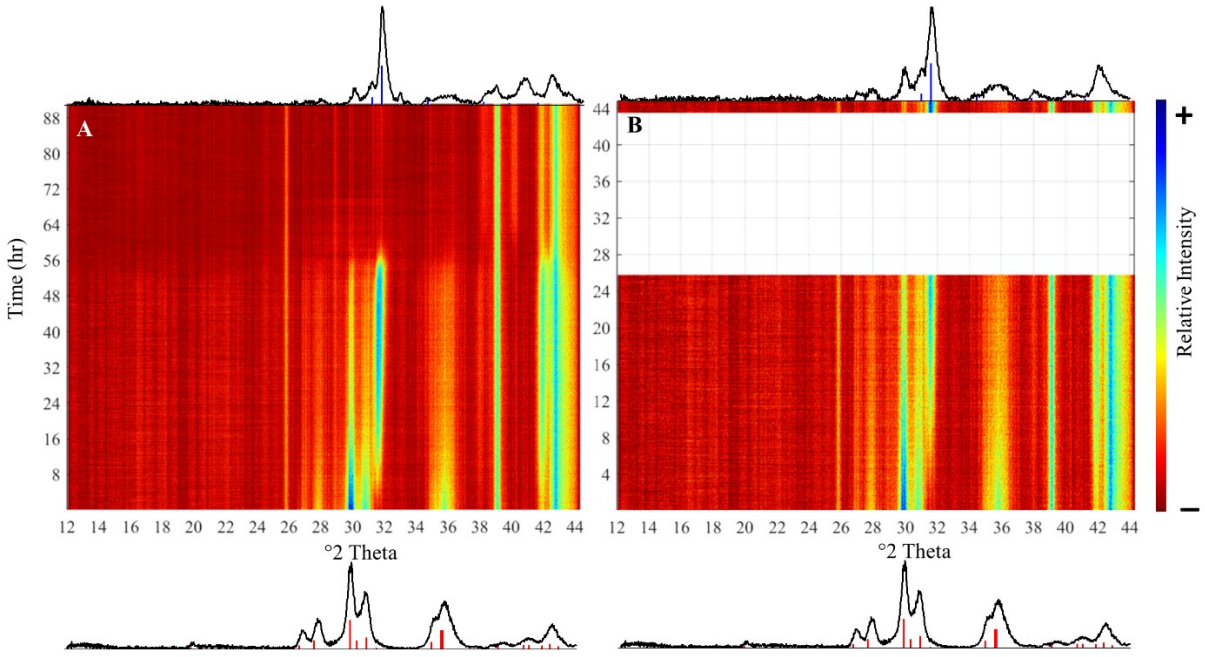

Figure S5. Full 2D waterfall plots for (A) **EXP IV** (sample fell from holder at ~56 hours) and (B) **EXP V** (gap in data collection due to temporary computer outage with no effect on *in situ* XRD reactor temperature or pressure).

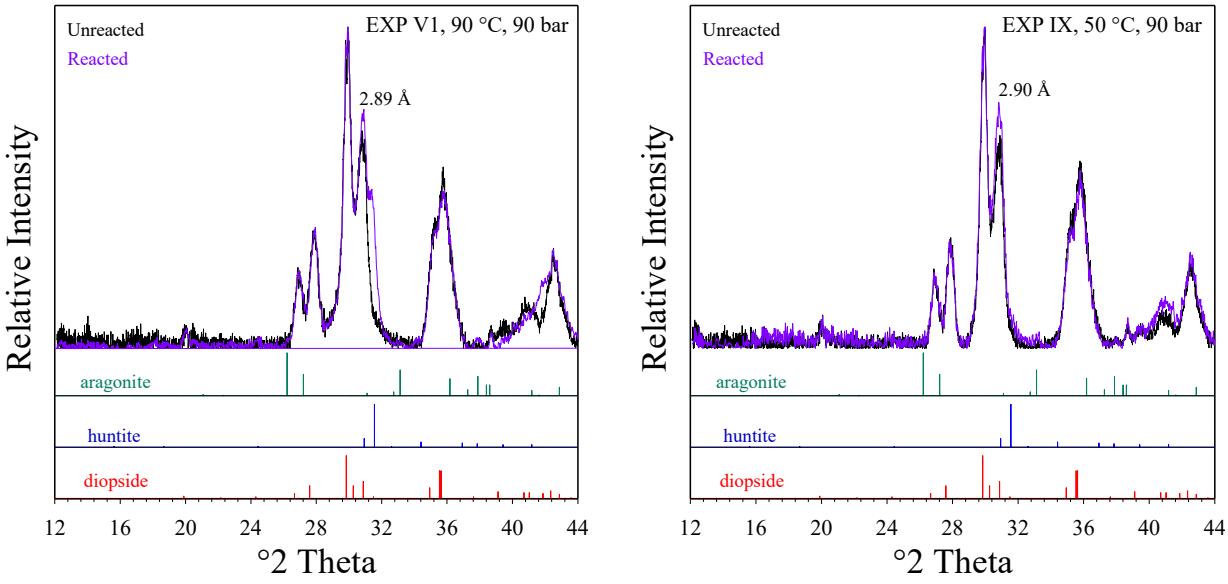

Figure S6. Overlap of initial and final *ex situ* patterns for **EXP VI** and **EXP IX**.

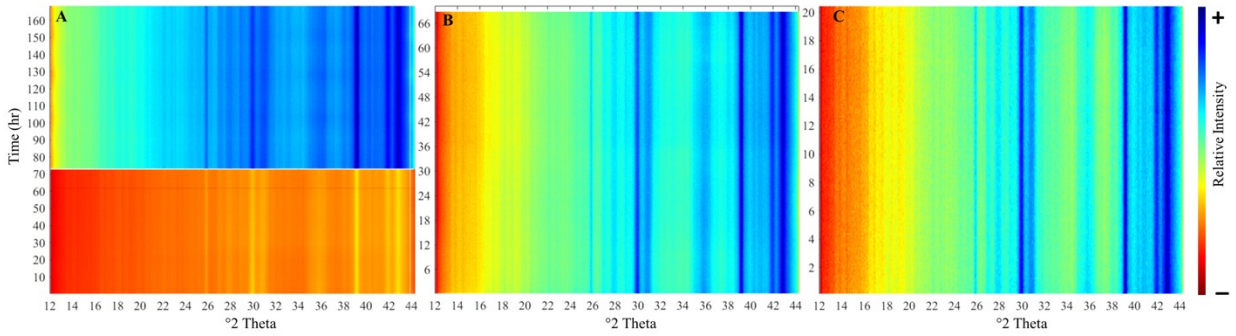

Figure S7. 2D waterfall plots for (A) **EXP VII**, (B) **EXP VIII**, and (C) **EXP IX** at 50 ˚C/90 bar. Panel A scan time was changed to 500 s after 72 hr.

Supplemental Reference

1. E. Tajuelo Rodriguez, L. M. Anovitz, C. D. Clement, A. J. Rondinone and M. C. Cheshire, Facile emulsion mediated synthesis of phase-pure diopside nanoparticles, *Sci. Rep.*, 2018, **8**, 3099.