

Supplementary Material

S1. Fresh Olivine Characterization

Figure S1 gives the particle size distribution of milled fresh olivine, determined by wet laser diffraction. A total of 86 vol. % of the material has a particle size below 80 μm and the average mean diameter $D\{4,3\}$ is equal to 34.80 μm .

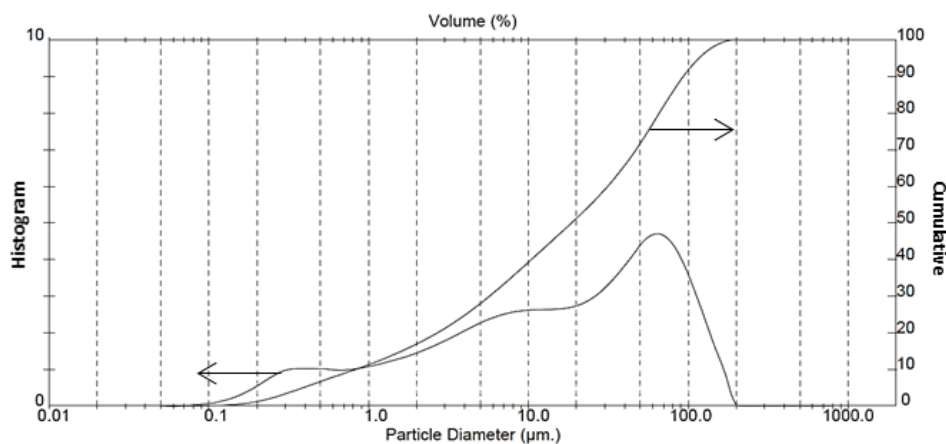


Figure S1. Particle size distribution by volume of milled fresh olivine; histogram (**left axis**) and cumulative (**right axis**).

The morphology of the milled fresh olivine particles was observed under scanning electron microscopy, and can be seen in Figure S2.

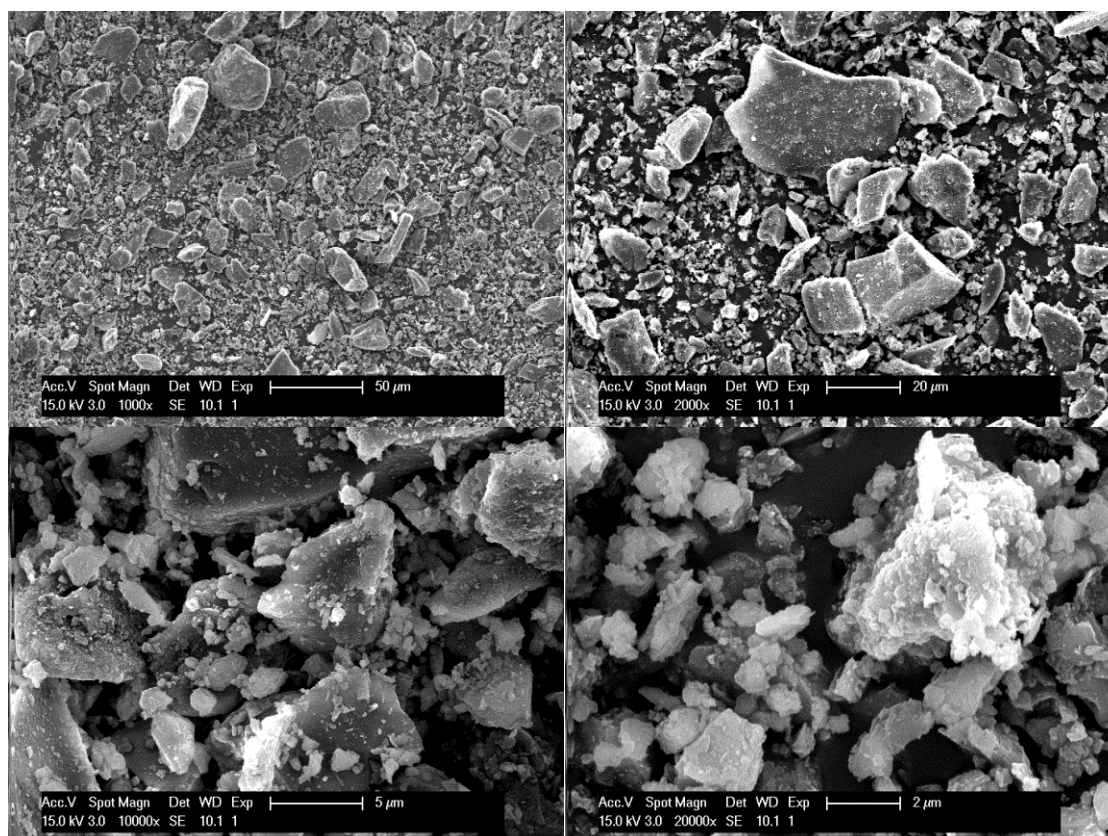


Figure S2. SEM images of milled fresh olivine.

A small surface of a polished fresh olivine sample (pelletized and embedded in resin) was fully mapped by EPMA to give the distribution of elements in the material. The mapping of the various elements can be seen in Figure S3. As can be expected, magnesium and silica are present all over the material as forsterite is the major mineral in the material. Iron, on the other hand, is more concentrated in certain particles, meaning that fayalite is not evenly dispersed. This can also be seen in Figure S4. Chromium, calcium and aluminum are also concentrated in certain particles and not evenly dispersed throughout the material. Nickel seems to be everywhere in the olivine. This would indicate that it replaces magnesium in the magnesium silicate structure to form a magnesium-nickel silicate ((Mg,Ni)₂SiO₄). There are also small particles that are highly concentrated (shown as white) in nickel.

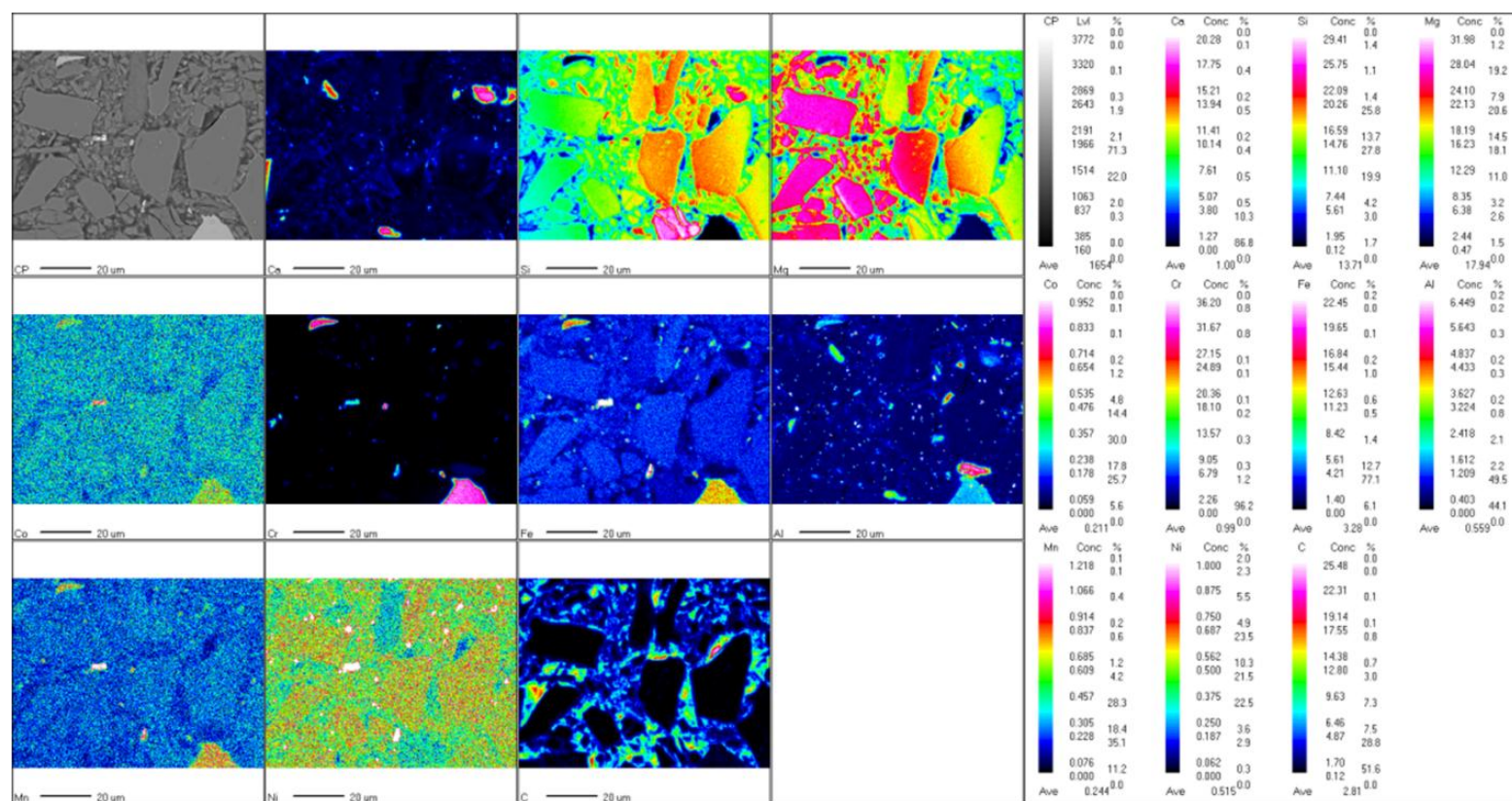
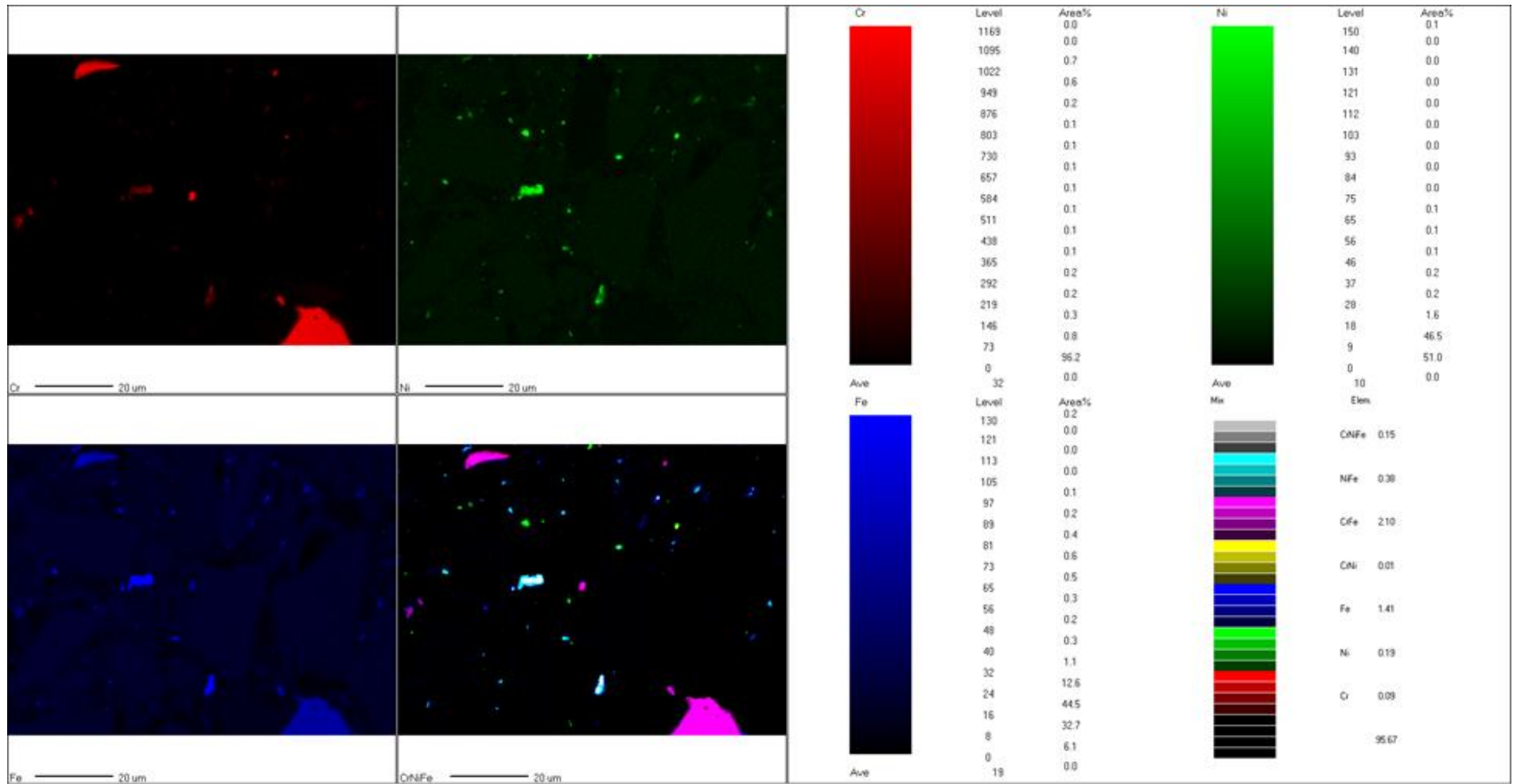


Figure S3. EPMA elemental concentration mapping of fresh olivine; “CP” image is backscattered electron analysis for sample visualization; color gradients in Mg and Si maps are due to curvature artifact that occurs at these relatively low magnifications.



S2. Preliminary Acid Leaching

Figure S5 shows the leaching results of the preliminary testing where seven different organic acids, at the same 2 N concentration, were used to leach fresh and carbonated olivine (2 g/100 mL for 24 h). Based on these results, citric, lactic and formic acids were chosen for the extensive testing presented in the main text.

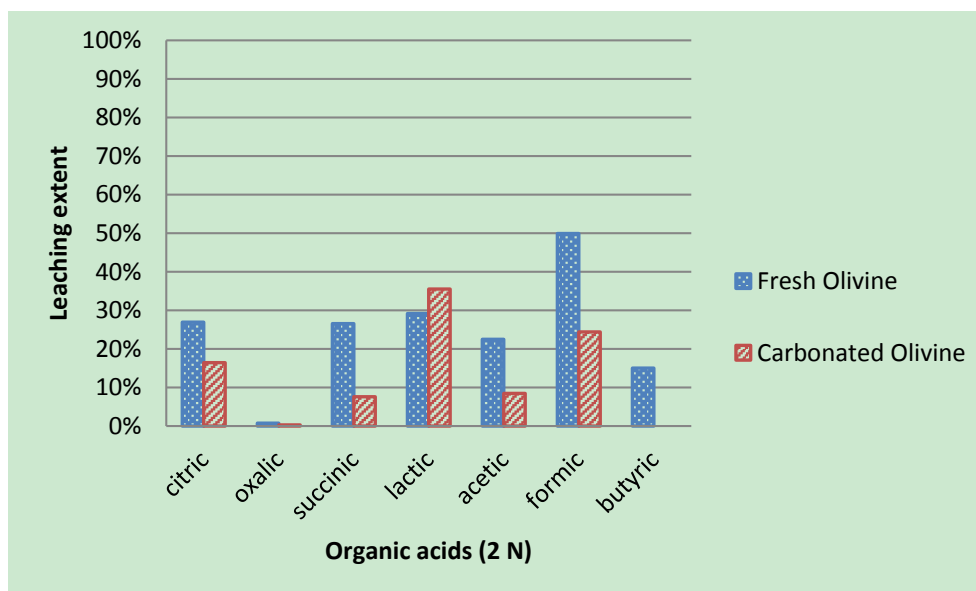


Figure S5. Nickel leached by various organic acids for fresh and fully carbonated olivine.

S3. Olivine Carbonation

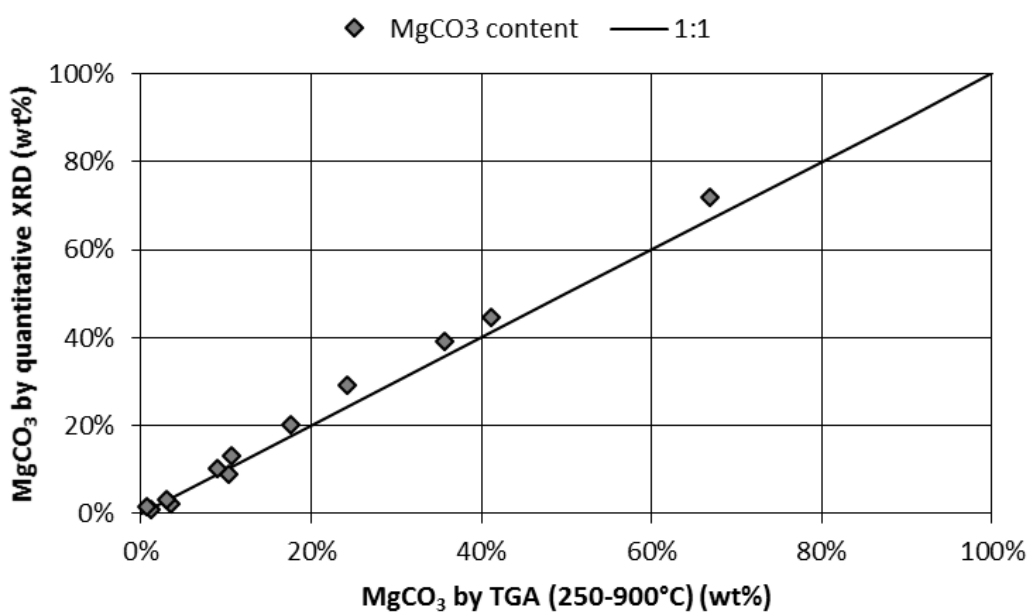


Figure S6. Comparison of magnesite content determination in carbonated olivine (processed under various conditions) by TGA and quantitative XRD.

Table S1. Carbonation extent for each condition and statistical data; shaded cells are replicates.

Carbonation Conditions (Time, Temp, Solids, Additives)	Conversion Extent (%)	TGA Variance (±)	Replicate Average Extent (%)	Replicate Variance (±)
4 h/Room T/50g/1 M NaCl + 0.64 M NaHCO ₃	2.1%	-		
4 h/150 °C/50 g/no salts	1.2%	-		
4 h/150 °C/50 g/1 M NaCl	0.7%	0.2%		
4 h/150 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	3.6%	-		
4 h/150 °C/50 g/1 M NaCl + 2.5 M NaHCO ₃	9.3%	0.2%		
4 h/150 °C/50 g/0.64 M NaHCO ₃	3.1%	-		
4 h/200 °C/50 g/no salts	3.8%	0.5%		
4 h/200 °C/50 g/1 M NaCl	3.2%	-		
4 h/200 °C/50 g/2 M NaCl	4.3%	-		
4 h/200 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	11.0%	0.3%		
4 h/200 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	17.1%	0.2%	13.6%	2.4%
4 h/200 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	12.7%	0.1%		
4 h/200 °C/50 g/1 M NaCl + 2.5 M NaHCO ₃	6.1%	-		
4 h/200 °C/50 g/0.64 M NaHCO ₃	11.0%	0.2%		
4 h/200 °C/100 g/1 M NaCl	4.2%	-		
4 h/200 °C/200 g/1 M NaCl	13.6%	0.0%		
24 h/150 °C/50 g/1 M NaCl	4.1%	-		
24 h/150 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	19.5%	0.7%		
24 h/185 °C/50 g/1 M NaCl	7.6%	0.1%		
24 h/200 °C/50 g/1 M NaCl	17.8%	0.3%		
24 h/200 °C/50 g/1 M NaCl	12.5%	0.4%	12.8%	3.4%
24 h/200 °C/50 g/1 M NaCl	7.9%	0.1%		
24 h/200 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	27.9%	2.6%		
24 h/200 °C/50 g/1 M NaCl + 2.5 M NaHCO ₃	11.3%	0.3%		
24 h/200 °C/200 g/1 M NaCl	34.9%	0.1%		
48 h/200 °C/50 g/1 M NaCl	28.6%	0.1%		
48 h/200 °C/200 g/1 M NaCl + 0.64 M NaHCO ₃	52.1%	0.3%		
72 h/200 °C/50 g/1 M NaCl	52.4%	0.5%		
72 h/200 °C/50 g/1 M NaCl + 0.64 M NaHCO ₃	60.5%	3.8%		
72 h/200 °C/200 g/1 M NaCl	102.9%	0.1%	99.0%	3.9%
72 h/200 °C/200 g/1 M NaCl	95.0%	2.9%		

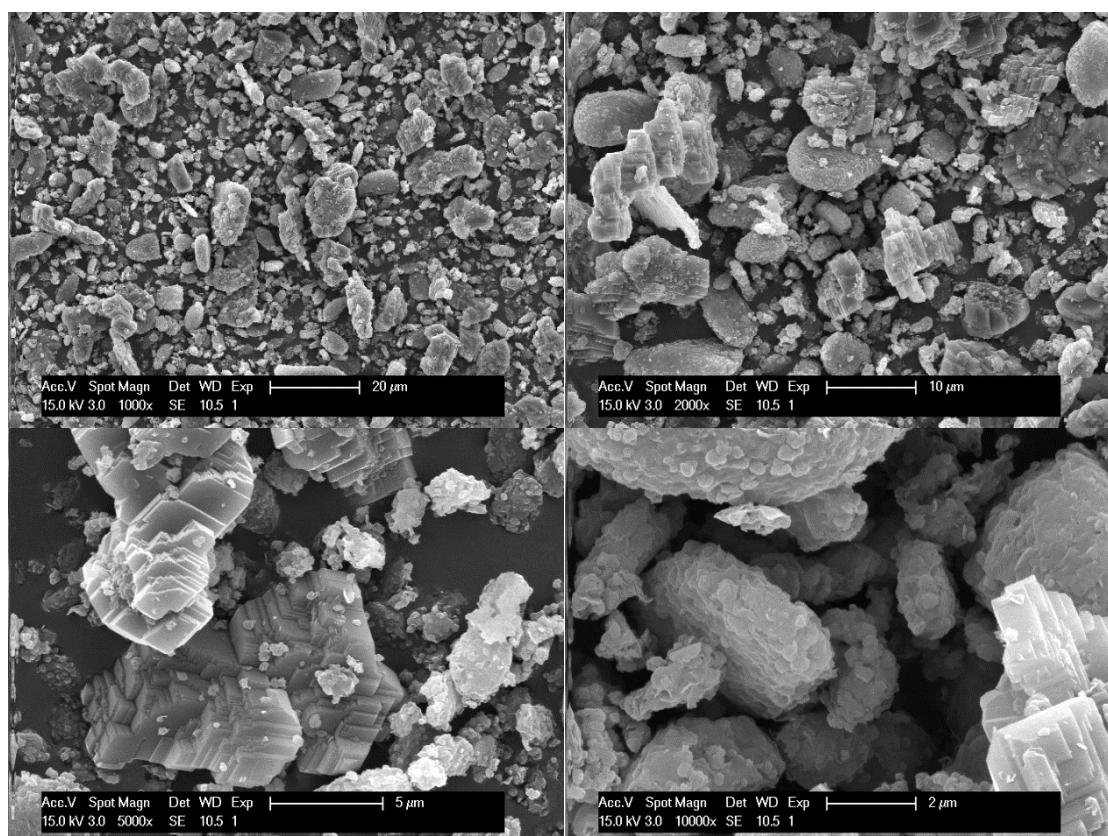


Figure S7. SEM images of partially carbonated olivine (72 h/200 °C/50 g/1 M NaCl; 52.4% $\pm$ 0.5% conversion extent).

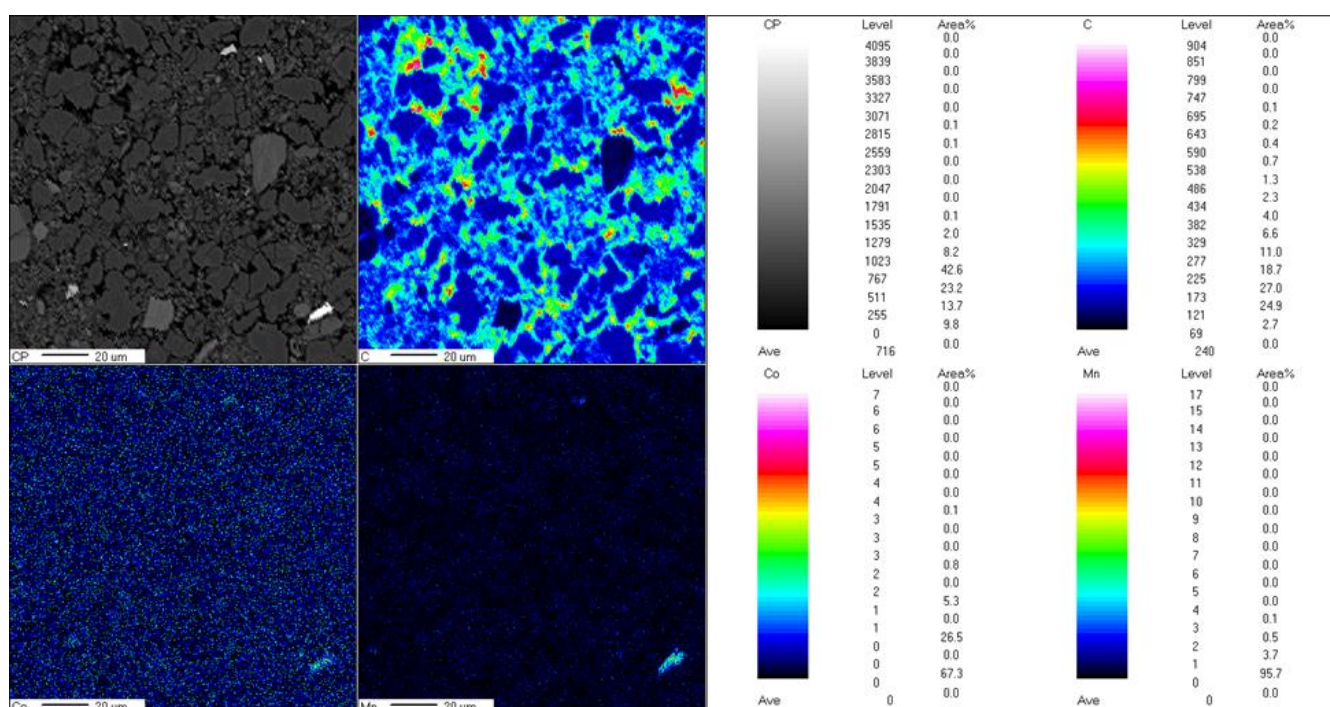


Figure S8. EPMA elemental concentration mapping of carbonated olivine (complementary to Figure 6 in the main text); “CP” image is backscattered electron analysis for sample visualization; concentration scale is relative to max/min levels; areas of highest carbon concentrations correspond to embedding resin.

S4. Acid Leaching

Tables S2–S5 show the variance of the leaching data presented in Figures 9 and 10 of the main text.

Table S2. Variance ($\pm$) of inorganic acid leaching results for fresh olivine.

Acid	Normality	Mg	Al	Si	Cr	Fe	Ni
HCl	0.02 N	0.7%	0.8%	0.1%	0.6%	1.0%	3.6%
	0.04 N	0.5%	4.0%	1.5%	0.2%	0.2%	2.1%
	0.08 N	3.6%	0.4%	0.0%	0.9%	3.5%	6.9%
	0.16 N	1.9%	2.7%	5.3%	0.6%	1.7%	1.0%
	0.32 N	5.1%	17.0%	3.5%	5.6%	0.1%	1.0%
	0.64 N	4.2%	12.2%	2.7%	5.1%	0.6%	0.4%
	1.28 N	1.2%	10.8%	3.2%	6.9%	3.2%	4.0%
	2.56 N	9.0%	4.3%	0.7%	0.1%	8.3%	8.7%
H₂SO₄	0.02 N	0.6%	2.5%	0.9%	0.1%	0.4%	1.7%
	0.04 N	0.4%	1.1%	1.6%	0.5%	0.2%	2.1%
	0.08 N	0.3%	2.0%	2.4%	0.4%	0.3%	3.0%
	0.16 N	3.6%	2.3%	5.8%	0.5%	3.3%	1.9%
	0.32 N	1.4%	10.3%	0.2%	4.6%	5.5%	6.2%
	0.64 N	5.3%	6.4%	2.7%	4.7%	8.9%	8.9%
	1.28 N	2.3%	16.0%	5.8%	5.1%	1.5%	1.4%
	2.56 N	0.0%	2.5%	0.8%	0.8%	0.4%	0.7%
HNO₃	0.02 N	0.0%	0.8%	0.5%	0.2%	0.3%	1.6%
	0.04 N	0.9%	3.9%	0.7%	0.4%	1.0%	1.4%
	0.08 N	2.1%	5.1%	1.8%	0.7%	2.3%	3.4%
	0.16 N	4.6%	2.6%	2.5%	0.8%	4.1%	4.8%
	0.32 N	0.9%	21.2%	2.2%	12.9%	1.7%	2.4%
	0.64 N	1.9%	8.7%	2.6%	11.1%	5.9%	5.6%
	1.28 N	2.0%	9.3%	0.1%	1.9%	4.8%	4.1%
	2.56 N	0.7%	2.7%	1.1%	4.3%	0.8%	2.8%

Table S3. Variance ($\pm$) of inorganic acid leaching results for carbonated olivine.

Acid	Normality	Mg	Al	Si	Cr	Fe	Ni
HCl	0.02 N	0.1%	0.6%	0.4%	0.0%	0.5%	0.1%
	0.04 N	0.6%	0.4%	0.4%	0.1%	0.2%	0.3%
	0.08 N	1.1%	0.9%	0.9%	0.1%	0.1%	0.5%
	0.16 N	3.6%	0.8%	0.8%	0.0%	2.0%	2.3%
	0.32 N	7.9%	0.5%	5.6%	0.1%	9.5%	7.1%
	0.64 N	9.2%	1.2%	3.8%	1.6%	11.8%	5.9%
	1.28 N	2.2%	1.0%	7.6%	1.6%	1.8%	2.5%
	2.56 N	8.9%	5.1%	1.6%	17.2%	13.5%	15.7%
H₂SO₄	0.02 N	0.3%	0.7%	1.0%	0.0%	0.4%	0.8%
	0.04 N	0.9%	0.7%	0.9%	0.1%	1.0%	1.1%
	0.08 N	0.6%	0.8%	0.9%	0.1%	0.7%	0.6%
	0.16 N	0.9%	1.1%	0.5%	0.1%	0.9%	0.9%
	0.32 N	1.5%	1.3%	4.5%	0.3%	0.2%	1.2%
	0.64 N	1.6%	2.9%	8.9%	0.6%	0.1%	0.0%
	1.28 N	1.7%	5.7%	0.8%	0.2%	0.0%	0.2%
	2.56 N	0.3%	2.3%	1.1%	1.5%	0.9%	1.8%
HNO₃	0.02 N	0.7%	0.5%	0.7%	0.0%	0.2%	0.5%
	0.04 N	1.0%	0.3%	0.6%	0.0%	0.4%	0.7%
	0.08 N	1.9%	0.5%	0.3%	0.0%	2.0%	1.6%
	0.16 N	2.2%	0.8%	0.6%	0.1%	3.2%	2.2%
	0.32 N	0.4%	2.4%	6.8%	0.9%	1.3%	0.2%
	0.64 N	7.7%	1.7%	9.0%	0.5%	5.4%	4.6%
	1.28 N	3.4%	2.6%	6.2%	0.2%	0.3%	0.1%
	2.56 N	5.4%	6.2%	0.7%	1.6%	5.7%	5.0%

Table S4. Variance ($\pm$) of organic acid leaching results for fresh olivine.

Acid	Normality	Mg	Al	Si	Cr	Fe	Ni
Formic	0.25 N	0.1%	1.6%	0.6%	0.1%	1.1%	3.9%
	0.5 N	0.2%	1.3%	1.0%	0.1%	2.2%	4.0%
	1 N	2.1%	2.6%	2.6%	0.2%	1.3%	2.9%
	2 N	0.8%	1.2%	0.3%	0.1%	3.4%	6.7%
	4 N	0.6%	2.2%	0.3%	0.2%	3.7%	7.4%
Citric	0.25N	3.2%	2.2%	2.5%	0.0%	1.6%	0.9%
	0.5 N	4.8%	3.3%	3.2%	0.4%	3.0%	0.8%
	1 N	4.8%	3.0%	2.6%	0.0%	3.0%	0.2%
	2 N	5.4%	3.7%	2.1%	0.1%	3.3%	0.8%
	4 N *	-	-	-	-	-	-
Lactic	0.25 N	1.8%	2.5%	1.9%	0.0%	0.7%	0.8%
	0.5 N	0.9%	1.2%	0.9%	0.1%	0.4%	2.5%
	1 N	0.8%	0.1%	0.2%	0.4%	2.3%	4.5%
	2 N	0.7%	0.1%	0.7%	0.1%	2.3%	4.6%
	4 N	0.2%	1.7%	1.0%	0.5%	2.3%	4.8%

* exceeds citric acid solubility.

Table S5. Variance ($\pm$) of organic acid leaching results for carbonated olivine.

Acid	Normality	Mg	Al	Si	Cr	Fe	Ni
Formic	0.25 N	1.8%	0.2%	0.3%	0.3%	2.2%	1.5%
	0.5 N	4.8%	1.2%	0.2%	0.4%	5.3%	0.1%
	1 N	7.0%	1.4%	0.1%	0.2%	7.8%	2.6%
	2 N	16.1%	3.5%	0.6%	0.3%	17.4%	9.6%
	4 N	15.0%	6.4%	1.1%	0.7%	16.2%	9.5%
Citric	0.25 N	0.1%	1.0%	1.1%	0.7%	1.4%	2.3%
	0.5 N	1.8%	1.4%	1.1%	0.8%	0.8%	6.1%
	1 N	1.4%	0.9%	0.4%	0.7%	0.9%	7.5%
	2 N	3.2%	0.2%	0.5%	0.9%	2.8%	10.4%
	4 N *	-	-	-	-	-	-
Lactic	0.25 N	4.2%	0.9%	0.0%	0.4%	5.2%	1.9%
	0.5 N	5.1%	0.2%	0.4%	0.4%	5.9%	1.5%
	1 N	6.7%	0.3%	0.4%	0.6%	7.5%	3.3%
	2 N	4.8%	1.7%	0.3%	0.9%	5.7%	1.8%
	4 N	8.3%	1.7%	3.7%	0.5%	9.9%	5.2%

* exceeds citric acid solubility.