

1 **Supporting Information for:**

2 **Hydration of Dicalcium Silicate and Diffusion through Neo-**  
3 **Formed Calcium-Silicate-Hydrates at Weathered Surfaces**  
4 **Control the Long-Term Leaching Behaviour of Basic Oxygen**  
5 **Furnace (BOF) Steelmaking Slag**

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18 *Consists of 17 pages with 4 tables and 5 figures.*

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20 **SI Table S1.** Limits of detection for each element measured by ICP-OES.

Element	Limit of Detection (mmol L <sup>-1</sup> )
Na	0.2857
Mg	0.0178
K	0.0793
Fe	0.0075
Si	0.0416
Al	0.0312
P	0.0031
V	0.0008
Cr	0.0009
Mn	0.0011
Ti	0.0010
Ca	0.0204
Zn	0.0002
As	0.0004

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22 **SI Table S2.** Experimental conditions and solution concentrations, as determined by ICP-OES. DL = below detection limit.

Size Fraction	Day	pH	Conductivity ( $\mu\text{S}$ )	Na	Mg	K	Fe	Si	Al	P	V	Cr	Mn	Ti	Ca	Zn	As	
				(mmol L <sup>-1</sup> )														
Sand (0.5-1.0 mm)	0	10.9	53	DL	DL	DL	0.020	DL	DL	DL	DL	DL	0.004	DL	0.155	DL	DL	
		-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
		10.7	32	DL	DL	DL	0.008	DL	DL	DL	DL	DL	DL	0.002	DL	0.064	DL	DL
	1	11.8	790	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	1.087	DL	DL
		11.6	664	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.944	DL	DL
		11.7	725	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	1.182	DL	DL
	2	11.6	731	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	1.064	DL	DL
		11.5	575	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.816	DL	DL
		11.5	679	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.987	DL	DL
	5	11.0	186.1	DL	DL	DL	DL	0.091	DL	DL	DL	0.002	DL	0.001	DL	0.443	0.001	DL
		10.6	105.7	DL	DL	DL	DL	0.081	DL	DL	DL	0.002	DL	DL	DL	0.231	0.004	DL
		10.9	166.5	DL	DL	DL	DL	0.092	DL	DL	DL	0.002	DL	DL	DL	0.340	0.001	DL
	8	10.5	137.5	DL	DL	DL	DL	0.333	DL	DL	DL	0.011	DL	0.001	DL	0.420	DL	DL
		10.4	118.2	DL	DL	DL	DL	0.296	DL	DL	DL	0.007	DL	DL	DL	0.306	DL	DL
		10.4	119.7	DL	DL	DL	DL	0.292	DL	DL	DL	0.008	DL	DL	DL	0.415	DL	DL
	14	10.3	168.1	DL	DL	DL	DL	0.875	DL	DL	DL	0.024	DL	0.003	DL	0.683	DL	DL
		10.2	146.9	DL	DL	DL	DL	0.793	DL	DL	DL	0.020	DL	0.001	DL	0.511	DL	DL
		10.1	120.1	DL	DL	DL	DL	0.676	DL	DL	DL	0.018	DL	0.002	DL	0.431	DL	DL
	28	9.5	133.2	DL	DL	DL	0.028	1.161	DL	DL	DL	0.031	DL	0.008	DL	0.810	DL	DL
		9.8	127.5	DL	DL	DL	DL	1.100	DL	DL	DL	0.029	DL	DL	DL	0.536	DL	DL
		9.9	133	DL	DL	DL	DL	1.092	DL	DL	DL	0.029	DL	0.001	DL	1.163	DL	DL
	57	8.5	116.9	DL	0.019	DL	0.026	1.212	DL	DL	DL	0.036	DL	0.006	DL	0.932	DL	DL
		8.6	118.6	DL	DL	DL	DL	1.119	DL	DL	DL	0.034	DL	DL	DL	0.518	DL	DL
		8.7	122.9	DL	DL	DL	0.008	1.716	DL	0.004	0.051	DL	0.003	DL	0.867	DL	DL	
73	8.8	119.9	DL	DL	DL	DL	1.155	DL	DL	DL	0.037	DL	DL	DL	0.551	DL	DL	
	8.7	116.5	DL	DL	DL	DL	1.112	DL	DL	DL	0.036	DL	0.001	DL	0.547	DL	DL	
	8.7	114.2	DL	DL	DL	0.012	1.165	DL	DL	DL	0.037	DL	0.004	DL	0.655	DL	DL	

Size Fraction	Day	pH	Conductivity ( $\mu\text{S}$ )	Na	Mg	K	Fe	Si	Al	(mmol L <sup>-1</sup> )									
										P	V	Cr	Mn	Ti	Ca	Zn	As		
Gravel (2.0-5.0 mm)	0	10.3	24.9	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.038	DL	DL	
		10.2	20.59	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.026	DL	DL
		10.0	19.22	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.034	DL	DL
	1	10.8	156.9	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.386	DL	DL
		10.7	132	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.322	DL	DL
		10.7	134.4	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.313	DL	DL
	2	10.3	89.7	DL	DL	DL	DL	0.070	DL	DL	0.001	DL	DL	DL	DL	DL	0.221	DL	DL
		10.2	94.1	DL	DL	DL	DL	0.083	DL	DL	0.002	DL	DL	DL	DL	DL	0.239	DL	DL
		10.1	86.2	DL	DL	DL	DL	0.091	DL	DL	0.002	DL	DL	DL	DL	DL	0.210	DL	DL
	5	10.0	85.9	DL	DL	DL	DL	0.255	DL	DL	0.006	DL	DL	DL	DL	DL	0.233	DL	DL
		9.8	80.2	DL	DL	DL	DL	0.229	DL	DL	0.006	DL	DL	DL	DL	DL	0.219	DL	DL
		10.1	96.4	DL	DL	DL	DL	0.315	DL	DL	0.007	DL	DL	DL	DL	DL	0.281	0.000	DL
	8	9.8	91.1	DL	DL	DL	DL	0.398	DL	DL	0.009	DL	DL	DL	DL	DL	0.263	DL	DL
		9.7	87	DL	DL	DL	DL	0.370	DL	DL	0.009	DL	DL	DL	DL	DL	0.251	DL	DL
		9.8	92.7	DL	DL	DL	DL	0.456	DL	DL	0.010	DL	DL	DL	DL	DL	0.283	DL	DL
	14	9.6	94.2	DL	DL	DL	DL	0.714	DL	DL	0.017	DL	DL	DL	DL	DL	0.332	DL	DL
		9.4	88.5	DL	DL	DL	DL	0.594	DL	DL	0.014	DL	DL	DL	DL	DL	0.301	DL	DL
		9.4	91.6	DL	DL	DL	DL	0.646	DL	DL	0.015	DL	DL	DL	DL	DL	0.314	DL	DL
	28	9.2	144.7	DL	DL	DL	DL	0.859	DL	DL	0.021	DL	DL	DL	DL	DL	0.405	DL	DL
		9.1	110.3	DL	DL	DL	DL	0.716	DL	DL	0.018	DL	DL	DL	DL	DL	0.384	DL	DL
		9.3	110.9	DL	DL	DL	DL	0.801	DL	DL	0.019	DL	DL	DL	DL	DL	0.407	DL	DL
	57	8.5	138	DL	DL	DL	DL	0.918	DL	DL	0.024	DL	DL	DL	DL	DL	0.620	DL	DL
		8.4	131.5	DL	DL	DL	DL	0.824	DL	DL	0.022	DL	DL	DL	DL	DL	0.562	DL	DL
		8.5	135.6	DL	DL	DL	DL	0.810	DL	0.003	0.022	DL	DL	DL	DL	DL	0.608	DL	DL
	73	8.5	133.3	DL	0.019	DL	DL	0.967	DL	DL	0.028	DL	DL	DL	DL	DL	0.636	DL	DL
		8.4	124.2	DL	DL	DL	DL	0.892	DL	DL	0.026	DL	DL	DL	DL	DL	0.572	DL	DL
		8.3	128.2	DL	DL	DL	DL	0.876	DL	DL	0.025	DL	DL	DL	DL	DL	0.581	DL	DL

Size Fraction	Day	pH	Conductivity ( $\mu\text{S}$ )	Na	Mg	K	Fe	Si	Al	P	V	Cr	Mn	Ti	Ca	Zn	As	
																		(mmol L <sup>-1</sup> )
Blocks (20 x 10 x 10 mm)	0	9.6	11.89	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	
		9.4	11.57	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL
	1	10.5	84.3	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.152	DL	DL
		10.3	66.7	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.114	DL	DL
	2	10.3	100.3	DL	DL	DL	DL	0.059	DL	DL	DL	0.001	DL	DL	DL	0.248	DL	DL
		10.1	80.5	DL	DL	DL	DL	0.049	DL	DL	DL	0.001	DL	DL	DL	0.183	DL	DL
	5	10.1	106.5	DL	DL	DL	DL	0.124	DL	DL	DL	0.003	DL	DL	DL	0.316	DL	DL
		9.9	95.9	DL	DL	DL	DL	0.106	DL	DL	DL	0.003	DL	DL	DL	0.279	0.001	DL
	8	9.7	85.2	DL	DL	DL	DL	0.178	DL	DL	DL	0.005	DL	DL	DL	0.249	DL	DL
		9.6	81.5	DL	DL	DL	DL	0.153	DL	DL	DL	0.004	DL	DL	DL	0.231	DL	DL
	14	9.5	73.3	DL	DL	DL	DL	0.271	DL	DL	DL	0.009	DL	DL	DL	0.231	DL	DL
		9.4	72.8	DL	DL	DL	DL	0.240	DL	DL	DL	0.008	DL	DL	DL	0.230	DL	DL
	28	9.3	77.9	DL	DL	DL	DL	0.366	DL	DL	DL	0.014	DL	DL	DL	0.263	DL	DL
		9.2	79.2	DL	DL	DL	DL	0.321	DL	DL	DL	0.011	DL	DL	DL	0.258	DL	DL
	57	8.3	101	DL	DL	DL	DL	0.479	DL	DL	DL	0.021	DL	DL	DL	0.351	DL	DL
		8.2	94.2	DL	DL	DL	DL	0.415	DL	DL	DL	0.017	DL	DL	DL	0.293	DL	DL
	73	8.0	95.6	DL	DL	DL	DL	0.508	DL	DL	DL	0.024	DL	DL	DL	0.379	DL	DL
		7.6	87.3	DL	DL	DL	DL	0.434	DL	DL	DL	0.019	DL	DL	DL	0.309	DL	DL
	Pre-weathered Block (20 x 10 x 10 mm)	0	9.0	10.28	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL
		1	9.1	18.45	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL
2		9.0	20.53	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	
5		8.9	28.5	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.042	DL	DL	
8		8.7	33.6	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	DL	0.062	DL	DL	
14		8.6	43	DL	DL	DL	DL	DL	DL	DL	DL	0.001	DL	DL	DL	0.115	DL	DL
28		8.6	63.6	DL	DL	DL	DL	0.064	DL	DL	DL	0.002	DL	DL	DL	0.198	DL	DL
57		7.8	91	DL	DL	DL	DL	0.084	DL	DL	DL	0.003	DL	DL	DL	0.282	DL	DL
73	6.7	85.7	DL	DL	DL	DL	0.076	DL	DL	DL	0.003	DL	DL	DL	0.248	DL	DL	

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29 **SI Table S3.** Average phase composition determined by SEM-EDS spot analysis performed on the  
 30 unreacted  $\text{Ca}_2\text{SiO}_4$  phase within BOF slag particles and the Ca-Si-H phase that replaces  $\text{Ca}_2\text{SiO}_4$  in the  
 31 surface alteration zone.

Element	A. $\text{Ca}_2\text{SiO}_4$	B. Ca-Si-H	Enrichment factor (B/A)
	n = 17 Mol % $\pm 1\sigma$	n = 89 Mol % $\pm 1\sigma$	
O	56.4 $\pm 2.7$	55.6 $\pm 9.2$	1.0
Mg	0.13 $\pm 0.09$	0.41 $\pm 0.77$	3.2
Al	0.20 $\pm 0.14$	0.70 $\pm 0.33$	3.4
Si	11.8 $\pm 0.65$	17.7 $\pm 5.9$	1.5
P	1.41 $\pm 0.08$	4.45 $\pm 1.46$	3.2
S	n.d.*	0.16 $\pm 0.14$	-
Cl	n.d.*	0.12 $\pm 0.07$	-
Ca	27.4 $\pm 1.4$	16.1 $\pm 5.1$	0.6
Sc	0.19 $\pm 0.04$	0.12 $\pm 0.08$	0.6
Ti	0.14 $\pm 0.10$	0.50 $\pm 0.28$	3.6
V	0.22 $\pm 0.22$	0.28 $\pm 0.35$	1.3
Mn	0.05 $\pm 0.02$	0.25 $\pm 0.44$	5.5
Fe	0.50 $\pm 0.11$	2.43 $\pm 2.43$	5.0
W	0.06 $\pm 0.01$	0.13 $\pm 0.04$	2.2
<i>Total</i>	<i>98.5</i>	<i>99.0</i>	

32 \*not detected.

33 **SI Table S4.** Chemical composition of phases in the surface alteration zone as a function of distance from the surface. Measured by SEM-EDS.

Size Fraction	Phase	Distance from surface (μm)	O	Mg	Al	Si	P	S	Cl	Ca	Sc	Ti	V	Mn	Fe	Lu	W
			mol %														
Sand (0.5-1.0 mm)	Ca-Si-H	0	67.25	0.21	0.69	15.30	3.26	0.04	0.12	10.81	0.07	0.40	0.12	0.14	1.52	-	0.09
	Ca-Si-H	3	49.11	0.24	0.78	24.01	3.88	0.06	0.12	17.92	0.12	0.74	0.23	0.15	2.43	0.04	0.18
	Ca-Si-H	5	38.82	0.24	0.83	31.47	3.84	ND	0.20	19.61	0.12	1.04	0.24	0.17	3.21	ND	0.23
	Ca-Si-H	10	49.61	0.33	0.72	26.09	2.62	0.18	0.17	15.13	ND	0.89	0.33	0.21	3.65	ND	0.07
	Ca-Si-H	15	43.79	0.29	0.86	27.52	3.63	0.07	0.20	18.03	0.13	0.63	0.20	0.26	4.26	-	0.18
	Ca-Si-H	18	59.47	0.36	0.65	21.80	3.52	0.07	0.12	11.70	0.08	0.38	0.10	0.10	1.53	0.00	0.12
	Ca-Si-H	25	39.27	0.26	0.52	23.23	2.86	ND	0.17	14.63	0.10	0.59	0.23	0.14	2.20	ND	0.15
	Ca-Si-H	29	55.93	0.41	0.56	24.75	3.93	ND	0.11	11.79	0.11	0.40	0.12	0.08	1.67	ND	0.14
	Ca-Si-H	34	57.13	0.42	0.50	23.12	4.00	ND	0.11	12.35	ND	0.41	0.10	0.11	1.64	ND	0.13
	Ca-Si-H	40	55.98	0.39	0.46	23.83	3.98	ND	0.15	12.86	ND	0.38	0.14	0.08	1.58	ND	0.16
	Ca-Si-H	46	65.06	0.51	0.58	20.14	2.76	0.16	0.08	8.71	0.07	0.28	0.08	0.09	1.36	0.01	0.08
	Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	52	62.32	0.19	0.19	11.32	1.39	0.06	ND	23.50	0.14	0.07	0.09	0.06	0.54	ND	0.05
	Ca <sub>2</sub> SiO <sub>4</sub>	60	57.38	ND	0.14	12.08	1.50	ND	ND	27.91	0.17	0.09	0.11	0.04	0.53	ND	0.05
	Ca <sub>2</sub> SiO <sub>4</sub>	63	52.07	0.23	ND	10.99	1.32	ND	ND	25.70	0.26	0.07	0.19	0.04	0.53	0.00	0.06
	Ca <sub>2</sub> SiO <sub>4</sub>	65	50.47	ND	0.16	10.66	1.30	ND	ND	24.82	0.24	0.07	0.16	0.06	0.54	ND	ND
	Ca-Si-H	0	55.84	0.30	0.65	23.12	4.41	ND	0.22	12.74	ND	0.39	0.12	0.17	1.87	0.00	0.11
Ca-Si-H	3	44.99	0.35	0.55	26.44	4.82	ND	0.19	18.88	0.10	0.56	0.15	0.17	2.58	ND	0.23	
Ca-Si-H	6	36.24	0.50	0.67	31.73	5.29	ND	0.22	19.66	0.16	0.61	0.15	0.43	4.15	ND	0.19	
Ca-Si-H	10	37.81	1.26	0.62	28.67	4.53	0.19	0.21	15.63	0.10	0.51	0.13	1.26	8.79	ND	0.20	
Ca-Si-H	16	47.30	0.47	0.69	30.12	3.65	ND	0.19	12.88	0.08	0.62	0.15	0.34	3.22	ND	0.16	
Ca-Si-H	20	56.86	0.34	0.82	26.10	2.63	0.12	0.17	9.76	0.09	0.57	0.10	0.17	2.18	0.00	0.08	

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SI Table S4. Continued.

Size Fraction	Phase	Distance from surface (µm)	O	Mg	Al	Si	P	S	Cl	Ca	Sc	Ti	V	Mn	Fe	Lu	W
			mol %														
Sand (0.5-1.0 mm)	Ca-Si-H	0	65.06	0.51	0.47	18.69	3.27	0.11	0.12	9.29	0.07	0.28	0.08	0.40	1.45	ND	0.10
	Ca-Si-H	5	58.32	0.72	0.59	24.48	3.20	ND	0.15	9.61	ND	0.44	0.09	0.11	2.16	ND	0.13
	Ca-Si-H	19	53.42	0.80	0.69	31.03	1.99	ND	0.17	8.50	0.56	0.07	0.04	0.10	2.55	ND	0.15
	Ca-Si-H	37	42.48	0.40	0.38	22.43	5.57	ND	0.10	24.71	0.16	0.48	0.24	0.22	2.67	ND	0.16
	Ca-Si-H	41	58.02	0.29	0.92	14.62	4.19	0.05	0.05	17.86	0.15	0.63	0.36	0.20	2.51	ND	0.11
	Ca-Si-H	49	45.29	0.24	0.26	10.14	2.92	ND	0.06	11.26	0.08	0.18	0.10	0.06	1.07	ND	0.11
	Ca-Si-H	52	44.59	0.35	0.81	16.15	5.75	0.07	0.10	26.08	0.15	0.56	0.55	0.31	4.26	ND	0.13
	Ca-Si-H	61	52.57	0.49	0.40	22.62	5.18	ND	0.10	16.32	0.11	0.33	0.13	0.10	1.51	ND	0.13
	Ca-Si-H	68	47.27	0.54	0.51	26.61	4.50	0.06	0.13	17.39	0.10	0.42	0.15	0.08	2.03	ND	0.17
	Ca-Si-H	71	61.57	0.80	0.41	20.39	3.30	0.07	0.07	11.66	0.08	0.25	0.09	0.05	1.12	ND	0.11
	Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	76	56.04	0.23	0.17	13.25	1.72	ND	ND	27.65	0.17	0.13	0.05	0.53	ND	ND	0.07
	Ca <sub>2</sub> SiO <sub>4</sub>	79	56.46	ND	0.13	12.34	1.51	ND	ND	28.62	0.19	ND	0.12	0.05	0.43	ND	0.07
	Ca <sub>2</sub> SiO <sub>4</sub>	82	57.33	ND	0.15	12.10	1.51	ND	ND	27.89	0.18	0.10	0.14	0.05	0.47	ND	ND
	Ca-Si-H	0	61.93	0.44	0.53	14.90	4.24	ND	0.12	13.26	ND	0.24	0.12	0.57	3.49	0.02	0.10
	Ca-Si-H	2	62.35	0.21	0.53	16.08	4.69	ND	0.06	14.11	0.11	0.24	0.13	0.13	1.26	-0.02	0.12
	Ca-Si-H	4	37.73	0.58	0.39	21.29	4.94	ND	0.06	19.54	ND	0.38	0.17	0.10	2.26	12.38	0.19
	Ca-Si-H	7	26.63	0.55	0.39	22.48	5.34	ND	0.08	21.70	ND	0.34	0.26	3.23	18.66	ND	0.19
	Ca-Si-H	11	37.51	0.19	0.39	17.89	4.75	ND	0.08	18.40	0.12	0.26	0.15	0.14	1.43	-0.02	0.15
	Ca-Si-H	16	42.89	0.24	0.35	23.89	5.84	ND	0.10	23.45	0.12	0.42	0.20	0.16	2.12	ND	0.22
	Ca-Si-H	18	52.35	0.29	0.32	21.43	4.89	ND	0.06	17.99	0.08	0.32	0.19	0.17	1.74	ND	0.16
	Ca-Si-H	22	55.20	0.36	0.38	22.93	4.39	ND	0.09	14.32	0.07	0.33	0.12	0.10	1.57	ND	0.14
	Ca-Si-H	28	61.15	0.40	0.44	22.21	3.42	ND	0.12	10.32	0.08	0.28	0.09	0.08	1.31	ND	0.11
Ca-Si-H	33	53.74	0.25	0.30	18.19	2.67	ND	0.06	22.59	0.15	0.29	0.13	0.10	1.34	ND	0.14	
Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	36	58.90	0.23	0.23	15.47	2.22	ND	0.03	21.60	0.12	0.14	0.11	0.06	0.80	ND	0.10	
Ca <sub>2</sub> SiO <sub>4</sub>	38	58.29	0.06	0.10	12.00	1.44	ND	ND	27.24	0.15	0.07	0.08	0.05	0.46	ND	0.06	
Ca <sub>2</sub> SiO <sub>4</sub>	44	57.28	ND	0.11	12.23	1.48	ND	ND	28.12	0.19	ND	0.10	0.04	0.39	ND	0.07	

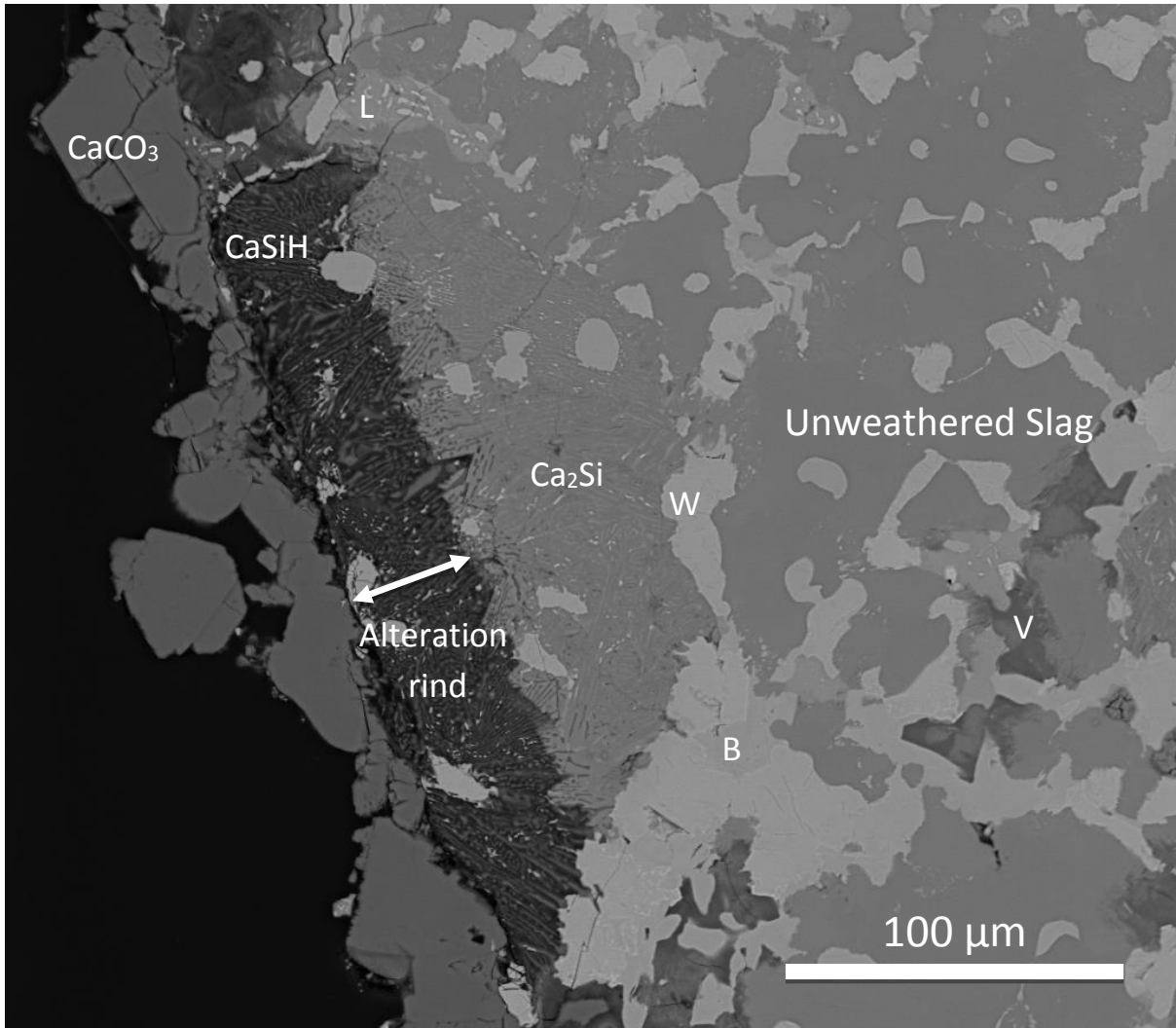


Size Fraction	Phase	Distance from surface (μm)	O	Mg	Al	Si	P	S	Cl	Ca	Sc	Ti	V	Mn	Fe	Lu	W
			mol %														
Gravel (2.0-5.0 mm)	Ca-Si-H	0	48.10	0.51	0.71	21.90	4.68	0.06	0.16	20.14	0.14	0.83	0.19	0.14	2.29	ND	0.16
	Ca-Si-H	3	52.03	0.45	0.54	19.30	3.76	ND	0.14	20.09	0.16	0.88	0.17	0.07	2.28	ND	0.15
	Ca-Si-H	5	41.22	0.55	0.48	23.08	4.60	0.09	0.20	25.00	0.17	1.06	0.21	0.10	3.05	ND	0.19
	Ca-Si-H	3	59.69	0.45	0.56	17.39	4.05	0.05	0.15	15.12	ND	0.41	0.12	0.08	1.81	ND	0.10
	Ca-Si-H	2	67.08	0.08	1.50	15.79	3.49	ND	0.15	8.21	0.06	1.01	0.12	0.13	2.24	ND	0.07
	Ca-Si-H	3	58.48	0.18	1.36	13.86	5.21	0.24	0.12	13.96	0.09	0.67	0.27	0.24	5.14	ND	0.09
	Ca-Si-H	2	51.91	0.37	0.92	10.38	2.90	0.04	0.08	7.02	0.05	0.41	0.05	0.17	1.82	ND	0.07
	Ca-Si-H	8	55.01	0.20	0.97	13.19	7.15	0.07	0.10	19.60	0.15	0.56	0.15	0.16	2.41	ND	0.15
	Ca-Si-H	12	61.21	0.14	0.67	8.82	6.46	0.08	0.07	18.21	0.11	0.35	0.17	0.29	3.04	ND	0.09
	Ca-Si-H	17	56.68	0.17	0.76	10.69	7.74	0.24	0.07	20.40	0.11	0.38	0.15	0.23	2.09	ND	0.13
	Ca-Si-H	23	63.36	0.20	0.48	7.60	6.16	0.31	0.06	19.20	0.11	0.30	0.12	0.19	1.75	ND	0.09
	Ca-Si-H	30	68.69	0.17	0.58	8.25	6.44	0.27	0.04	13.69	0.08	0.22	0.08	0.11	1.04	ND	0.09
	Ca-Si-H	39	58.98	0.14	0.62	10.96	8.03	0.16	0.09	18.32	0.14	0.36	0.11	0.17	1.57	ND	0.14
	Ca-Si-H	48	63.14	0.18	1.72	19.36	3.19	0.11	0.14	8.94	ND	0.75	0.13	0.16	2.06	ND	0.08
	Ca-Si-H	63	59.38	0.28	0.35	17.68	3.89	0.13	0.03	16.11	0.09	0.31	0.15	0.10	1.28	ND	0.12
	Ca-Si-H	66	69.45	0.36	0.38	14.47	3.95	0.11	0.02	10.07	0.07	0.16	0.08	0.05	0.66	ND	0.10
	Ca-Si-H	69	62.32	0.22	0.33	18.34	4.16	0.11	ND	13.01	0.08	0.18	0.11	0.07	0.86	ND	0.12
	Ca-Si-H	72	66.14	0.24	0.36	18.80	2.81	ND	ND	10.45	0.07	0.15	0.09	0.05	0.65	ND	0.10
	Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	75	71.34	0.07	ND	10.63	1.31	ND	ND	15.88	0.12	0.07	0.03	0.34	ND	ND	0.04
	Ca <sub>2</sub> SiO <sub>4</sub>	78	60.35	ND	0.19	11.95	1.35	ND	ND	25.41	0.17	ND	0.10	0.03	0.39	ND	0.05
Ca <sub>2</sub> SiO <sub>4</sub>	83	59.63	ND	0.17	12.00	1.34	ND	ND	26.01	0.18	ND	0.10	0.04	0.40	ND	0.06	
Ca <sub>2</sub> SiO <sub>4</sub>	90	57.99	0.09	0.20	12.06	1.47	ND	ND	26.64	0.31	ND	0.13	0.10	0.77	ND	0.05	

Size Fraction	Phase	Distance from surface (μm)	O	Mg	Al	Si	P	S	Cl	Ca	Sc	Ti	V	Mn	Fe	Lu	W
			mol %														
Gravel (2.0-5.0 mm)	Ca-Si-H	0	73.76	0.16	1.49	13.59	1.77	0.06	0.51	6.55	0.05	0.40	0.06	0.09	1.52	ND	ND
	Ca-Si-H	1.5	65.11	0.17	1.71	18.65	2.69	0.06	0.16	8.55	0.53	ND	0.07	0.11	2.11	ND	0.08
	Ca-Si-H	9	61.02	0.20	0.89	13.36	6.78	0.14	0.11	14.76	0.12	0.41	0.12	0.17	1.82	ND	0.12
	Ca-Si-H	19	57.59	0.15	0.85	15.37	6.37	0.08	0.07	16.15	0.12	0.56	0.11	0.15	2.27	ND	0.16
	Ca-Si-H	28	61.18	0.12	0.61	11.91	7.34	ND	0.13	16.18	0.10	0.36	0.12	0.13	1.60	ND	0.13
	Ca-Si-H	38	58.24	0.09	0.44	9.96	8.76	ND	0.11	20.02	0.15	0.30	0.15	0.18	1.50	ND	0.08
	Ca-Si-H	56	58.36	0.19	0.40	9.75	8.50	0.38	0.05	19.96	0.13	0.26	0.15	0.17	1.38	ND	0.13
	Ca-Si-H	69	64.58	0.36	0.75	13.30	4.83	0.32	0.08	12.37	ND	0.32	0.12	0.24	2.21	ND	0.10
	Ca-Si-H	69	48.90	7.30	0.71	11.70	3.53	0.22	0.08	10.11	ND	0.22	0.08	2.38	14.04	ND	0.08
	Ca-Si-H	88	61.38	0.28	0.50	15.86	5.72	0.54	0.12	13.06	0.10	0.29	0.10	0.14	1.45	ND	0.10
	Ca-Si-H	106	65.82	0.28	0.37	12.77	5.28	0.58	0.13	12.74	0.07	0.24	0.10	0.12	1.13	ND	0.11
	Ca-Si-H	125	56.46	0.34	0.42	17.26	5.76	0.45	0.07	16.75	0.12	0.27	0.13	0.12	1.35	ND	0.15
	Ca-Si-H	144	57.79	0.38	0.39	17.90	5.40	0.32	0.04	15.45	0.11	0.28	0.14	0.08	1.22	ND	0.15
	Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	156	50.21	ND	0.11	10.05	1.34	ND	ND	22.77	0.13	0.08	0.09	0.05	0.43	ND	ND
	Ca <sub>2</sub> SiO <sub>4</sub>	163	57.31	ND	0.17	11.82	1.45	ND	ND	28.11	0.17	0.10	0.16	0.04	0.56	ND	0.06
	Ca <sub>2</sub> SiO <sub>5</sub>	181	57.22	ND	0.10	12.30	1.46	ND	ND	28.14	0.20	ND	0.10	0.04	0.40	ND	0.05
Ca <sub>2</sub> SiO <sub>6</sub>	206	57.16	ND	0.11	12.35	1.41	ND	ND	28.25	0.18	ND	0.11	0.02	0.37	ND	0.05	

Size Fraction	Phase	Distance from surface (μm)	O	Mg	Al	Si	P	S	Cl	Ca	Sc	Ti	V	Mn	Fe	Lu	W
			mol %														
Blocks (20 x 10 x 10 mm)	Ca-Si-H	0	70.66	0.13	1.47	11.50	3.73	0.06	0.07	9.73	0.05	0.73	0.10	0.08	1.57	ND	0.06
	Ca-Si-H	0	71.39	0.13	1.11	8.10	4.80	0.13	0.20	11.18	ND	0.65	0.30	0.19	1.74	ND	0.09
	Ca-Si-H	0	53.78	0.30	1.55	14.90	7.53	ND	0.19	18.02	ND	0.92	0.21	0.21	2.22	ND	0.12
	Ca-Si-H	1	56.62	0.39	0.72	14.84	4.74	0.11	0.17	19.38	ND	0.56	0.50	0.08	1.75	ND	0.13
	Ca-Si-H	2	63.87	0.46	0.56	13.48	4.37	ND	0.13	15.09	0.10	0.34	0.29	0.05	1.18	ND	0.07
	Ca-Si-H	2	51.53	0.71	0.97	13.99	4.39	0.53	0.28	21.52	0.14	0.72	0.65	0.41	4.01	ND	0.11
	Ca-Si-H	3	51.84	0.30	0.59	14.07	4.69	0.06	0.12	24.77	0.15	0.70	0.68	0.04	1.81	ND	0.17
	Ca-Si-H	8	56.87	0.33	0.62	14.09	4.80	ND	0.10	20.36	0.15	0.52	0.54	0.07	1.43	ND	0.13
	Ca-Si-H	15	54.30	0.24	0.87	15.19	4.47	ND	0.08	21.50	0.14	0.74	0.78	0.08	1.49	ND	0.12
	Ca-Si-H	24	42.35	0.16	1.41	16.55	3.41	0.12	0.08	28.32	0.15	1.62	1.48	0.24	3.98	ND	0.14
	Ca-Si-H	33	55.85	0.18	1.07	13.61	3.53	0.33	0.05	19.71	0.14	0.88	1.29	0.35	2.92	ND	0.10
	Ca-Si-H	42	50.52	0.24	0.86	16.16	3.89	ND	0.05	22.11	ND	1.04	1.30	0.55	3.13	ND	0.15
	Ca-Si-H	51	59.57	ND	0.46	11.84	1.64	ND	ND	24.50	0.16	0.41	0.69	0.05	0.62	ND	0.06
	Ca-Si-H	57	49.35	ND	0.93	13.02	3.27	0.04	ND	28.62	0.16	0.87	1.85	0.08	1.74	ND	0.09
	Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	60	56.90	ND	0.77	11.31	1.75	0.05	ND	26.85	0.15	0.39	0.86	0.05	0.86	ND	0.06
	Ca <sub>2</sub> SiO <sub>4</sub>	63	51.03	ND	0.57	10.17	1.31	ND	ND	26.39	0.14	0.26	0.77	0.04	0.46	ND	0.07
	Ca <sub>2</sub> SiO <sub>5</sub>	72	55.65	ND	0.54	11.10	1.40	ND	ND	29.20	0.16	0.37	0.76	0.05	0.72	ND	0.06
	Ca <sub>2</sub> SiO <sub>6</sub>	78	56.15	ND	0.19	12.12	1.37	ND	ND	29.00	0.17	0.15	0.31	0.04	0.46	ND	0.05

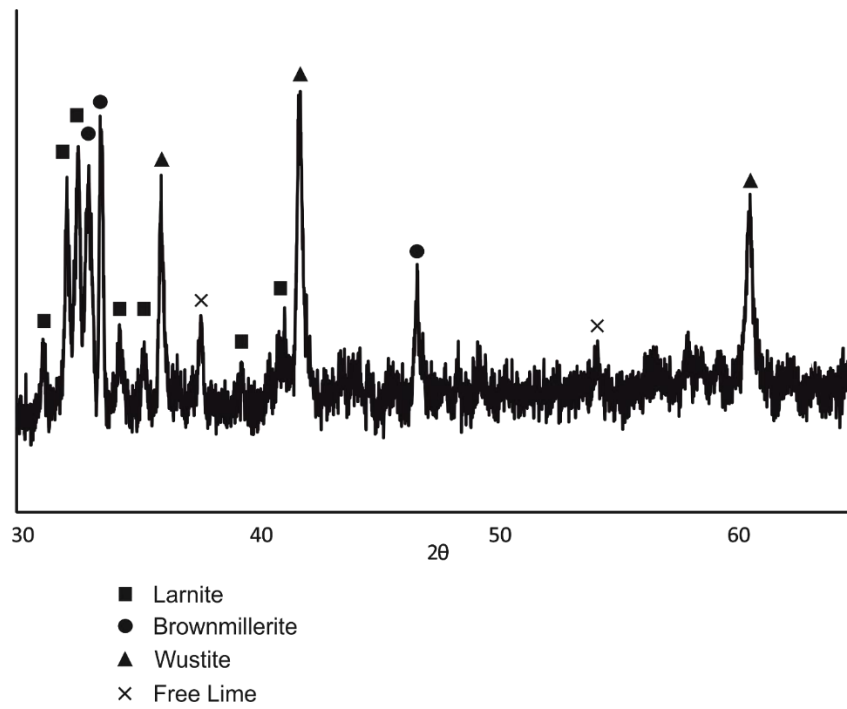
Size Fraction	Phase	Distance from surface (μm)	O	Mg	Al	Si	P	S	Cl	Ca	Sc	Ti	V	Mn	Fe	Lu	W
			mol %														
Blocks (20 x 10 x 10 mm)	Ca-Si-H	0	65.12	0.31	0.62	11.67	3.68	ND	0.29	13.43	ND	0.31	0.28	1.20	2.96	ND	0.11
	Ca-Si-H	1	72.27	0.34	0.50	10.21	3.49	ND	0.10	11.39	ND	0.28	0.20	0.12	0.93	ND	0.07
	Ca-Si-H	4	56.29	0.29	1.12	14.85	4.17	0.15	0.15	18.64	0.10	0.60	0.54	0.72	2.31	ND	0.09
	Ca-Si-H	9	63.47	0.25	0.63	13.16	4.16	ND	0.13	15.44	0.11	0.46	0.41	0.27	1.39	ND	0.11
	Ca-Si-H	18	60.92	0.18	0.59	13.08	4.30	ND	0.14	18.01	0.10	0.56	0.59	0.11	1.32	ND	0.10
	Ca-Si-H	22	56.35	0.19	0.94	16.01	3.70	ND	0.05	18.89	0.11	1.00	0.98	0.13	1.51	ND	0.13
	Ca-Si-H	25	51.85	0.21	1.05	16.49	3.64	0.05	0.05	21.36	0.14	1.22	1.46	0.20	2.13	ND	0.11
	Ca-Si-H/Ca <sub>2</sub> SiO <sub>4</sub>	28	58.13	ND	0.58	11.57	1.52	ND	0.03	25.99	ND	0.51	0.79	0.06	0.75	ND	0.05
	Ca <sub>2</sub> SiO <sub>4</sub>	32	56.37	ND	0.23	12.19	1.26	ND	ND	28.84	0.17	0.13	0.26	0.04	0.43	ND	0.05



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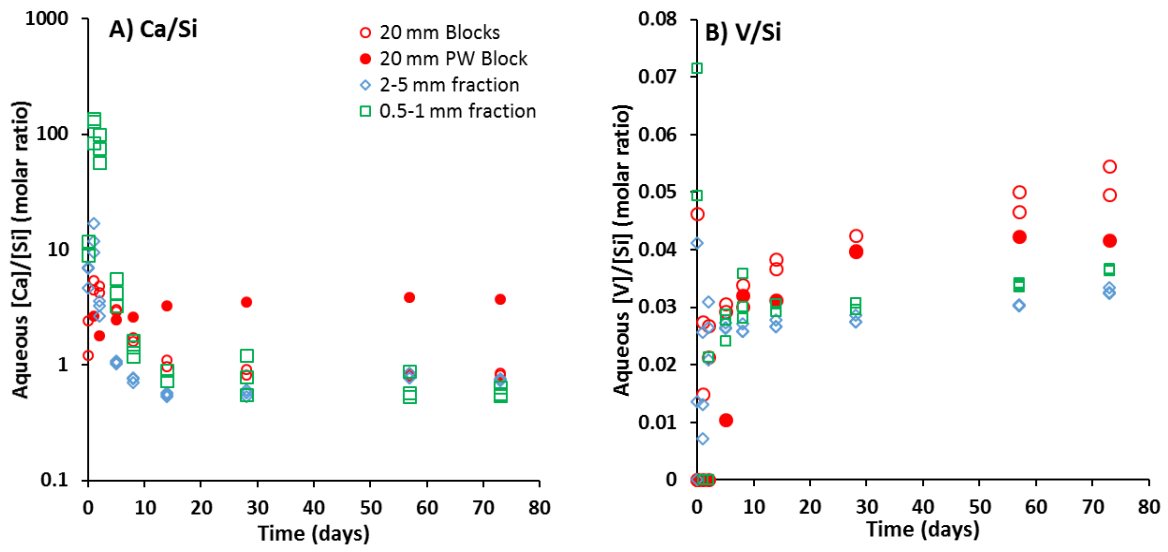
48 **SI Figure S1.** Example BSEI electron micrograph showing the primary ( $\text{Ca}_2\text{S}$  – larnite; B –  
 49 Brownmillerite; L – Lime; W – Wustite; V – void space) and secondary Ca-Si-H and  $\text{CaCO}_3$  phases  
 50 present at the surface of the aerobically weathered 20 mm BOF slag blocks after 6 months total  
 51 immersion. All phases were identified by EDS spot analysis of representative regions. Alteration  
 52 depths were defined as the changed surface region within the original volume of the slag particle  
 53 (presence of refractory phases allows good estimation of the original particle size); the thickness of  
 54 any  $\text{CaCO}_3$  layer was not included in the analysis.

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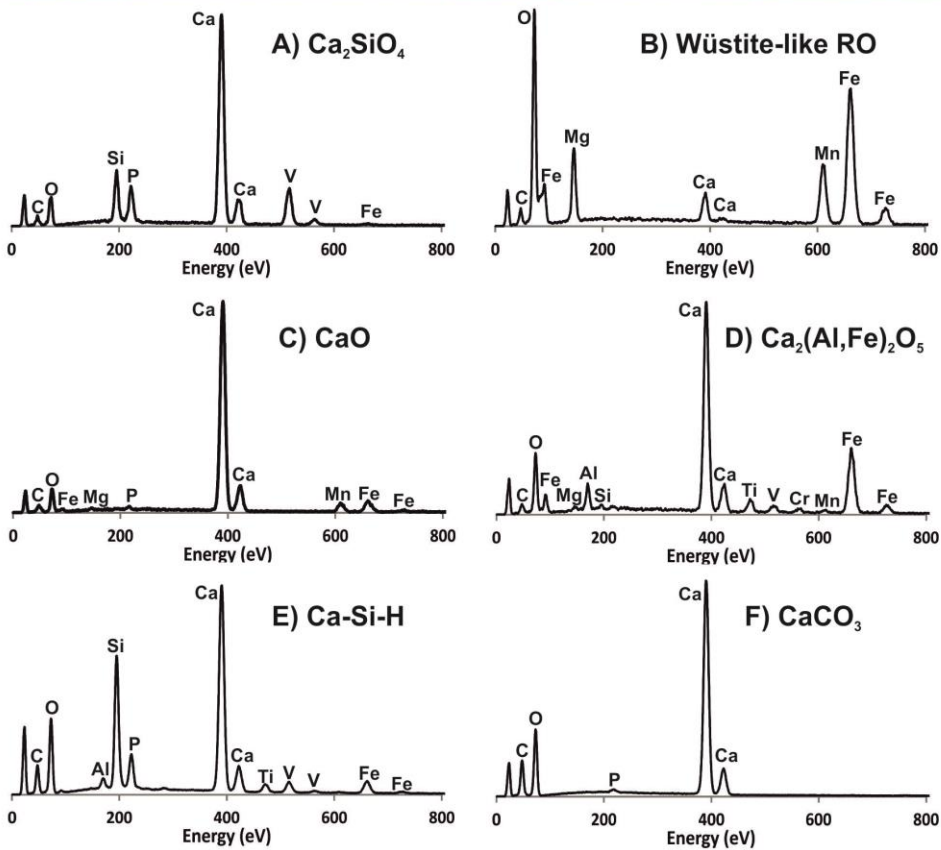
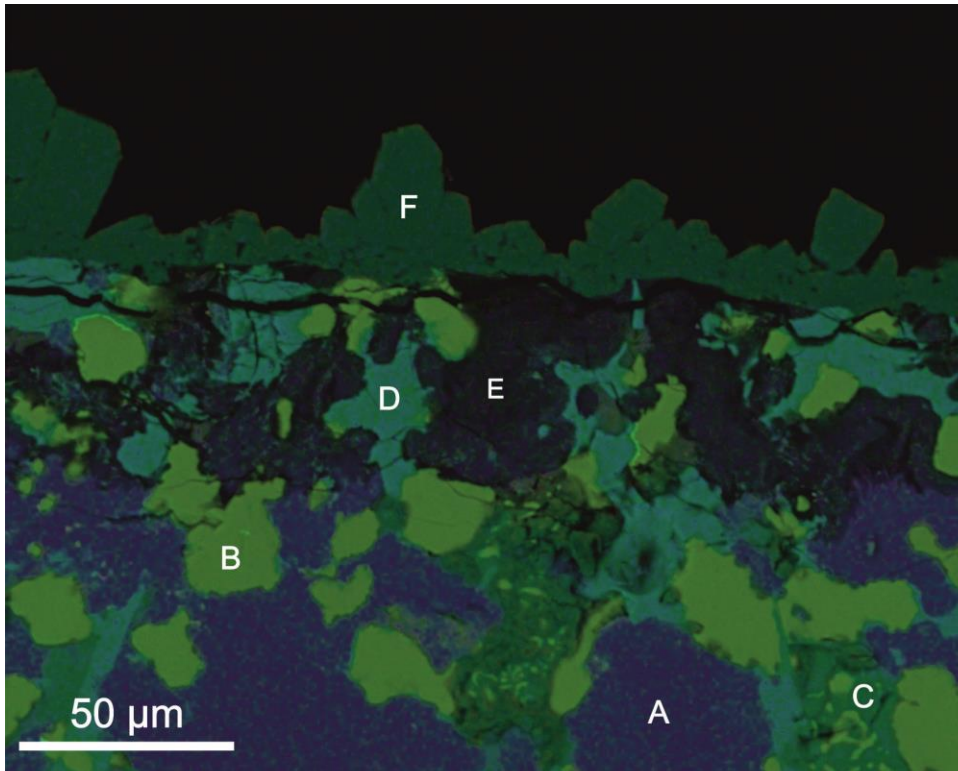
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**SI Figure S2.** XRD pattern collected from the crushed steel slag sample annotated with major phase peaks detected.



60 **SI Figure S3.** Elemental cross-plots showing; A) The relationships between aqueous [Ca] and [Si],  
 61 and; B) Aqueous [Si] and [V] in individual replicates during the leaching tests.

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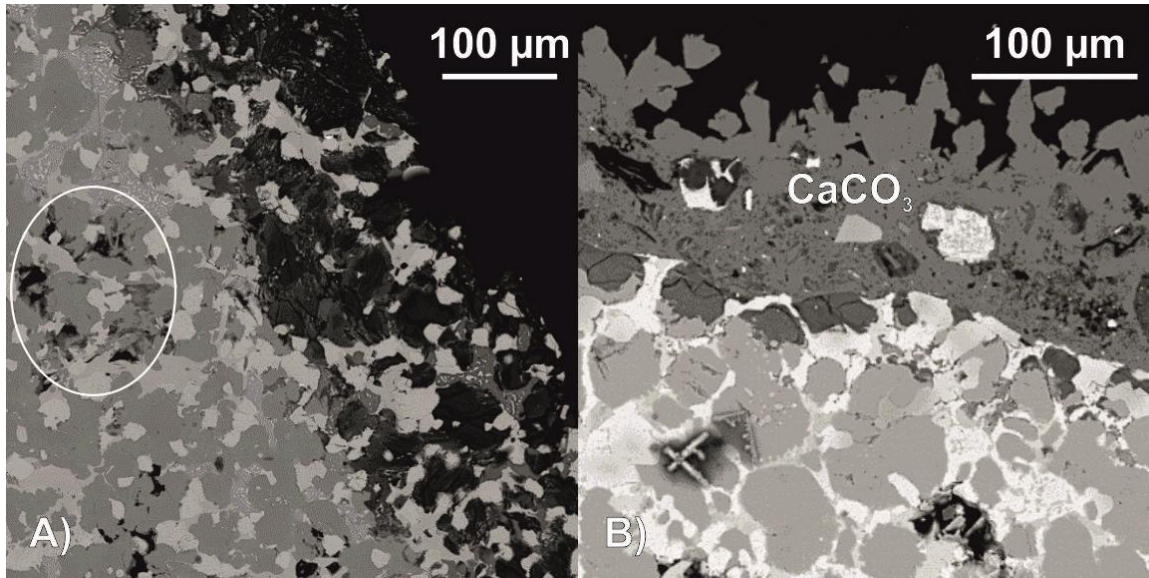
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**SI Figure S4.** Composite false colour SEM-EDS elemental map showing phase discrimination within the 6 month pre-weathered BOF slag block. A-D) Example EDS spectra collected from each of the 6 major phases detected with the slag; and E-F) Example EDS spectra from neo-formed phases present in the altered surface layer.





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69 **SI Figure S5.** BSE images of different sized BOF slag particles after leaching for 73 days; (a) Block  
70 showing possible Ca-Si-H formation within occasional voids remote from the block surface, and (b)  
71 Sand-sized fraction showing CaCO<sub>3</sub> crystals on the weathered surface.