

Table S0. Initial composition of Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ solids (mg/L) used in abiotic reduction tests. Experimental ratio is the molar ratio of Mg/X, where X =Fe or Al.

MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
(Mg)	(X)	Exp Ratio	(Mg)	(X)	Exp Ratio	(Mg)	(X)	Exp Ratio	(Mg)	(X)	Exp Ratio
24304	27923	0.87	24305	26982	0.91	24301	55845	0.44	24277	12378	1.96

Table S1. The average pH range, %Fe removed from solution, and %Mg released after 7 days for tests conducted at pH 8 for 7 days with 10 mM Fe(II)_(aq).

	MgFeCO ₃	MgAlCO ₃	MgFeSO ₄	MgAlSO ₄
pH range measured	8.0-8.9	7.8-8.5	8.2-8.9	7.5-7.7
% Fe removed	99	99	99	81
%Mg released [‡]	1.42	0.3	1.36	0.2

[‡] Mg released relative to the initial Mg solid concentration in Table S0.

Table S2. The average pH range, %Fe removed from solution, and %Mg released after 7 days for tests conducted at pH 8 with 0.5 mM Fe(II)_(aq).

	MgFeCO ₃	MgAlCO ₃	MgFeSO ₄	MgAlSO ₄
pH range measured	9.0-9.1	9.7-9.7	9.0-9.5	9.0-9.2
% Fe removed	99	99	99	99
%Mg released [‡]	0.21	0.01	0.44	0.02

[‡] Mg released relative to the initial Mg solid concentration in Table S0.

Table S3. The average pH range, %Fe removed from solution, and %Mg released after 7 days for tests conducted at pH 10 with 0.5 mM Fe(II)_(aq).

	MgFeCO ₃	MgAlCO ₃	MgFeSO ₄	MgAlSO ₄
pH range measured	9.2-9.7	9.9-10.0	9.2-9.5	9.7-10.1
% Fe removed	99	99	99	99
%Mg released [‡]	0.08	0.003	0.27	0.001

[‡] Mg released relative to the initial Mg solid concentration in Table S0.

Table S4. Leachability of Fe and Mg for individual tests conducted at pH 8 for 7 days with 10 mM Fe(II)_(aq). Time 0 days represents conditions before Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ solids were added. Concentration units for (Fe) and (Mg) are mg/L. All data reported here have a standard deviation (RSD) of ±10%.

Time (Days)	MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
	Test 1			Test 1			Test 1			Test 1		
	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)
0	5.5	749	0	5.5	749	0	5.5	749	0	5.5	749	0
1	7.8	1.95	239.0	7.8	20.0	50.4	8.7	2.19	248.5	7.6	330.1	46.3
2	8.9	0.66	448.1	8.3	0.65	64.2	8.7	0.61	276.2	7.5	299.3	49.9
4	8.9	0.04	404.3	8.5	0.43	60.5	9.0	0.2	285.2	7.5	184.4	61.4
7	8.8	0.15	411.9	8.6	0.5	51.5	9.0	0.1	282.5	7.5	131.3	47.5

Time (Days)	MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
	Test 2			Test 2			Test 2			Test 2		
	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)
0	5.5	749	0	5.5	749	0	5.5	749	0	5.5	749	0
1	8.2	4.9	355.0	7.8	21.2	65.1	7.7	17.5	233. 2	7.8	413.8	67.1
2	8.9	0.12	259.4	8.5	2.05	83.2 4	8.9	0.71	348. 1	7.8	322.3	63.6
4	8.9	0.12	262.2	8.6	0.74	85.4	8.9	0.32	376. 4	7.5	238.9	61.8
7	8.9	0.17	274.9	8.6	0.6	76.3	8.9	0.16	372. 2	7.5	149.9	46.3

Table S5. Leachability of Fe and Mg for individual tests conducted at pH 8 for 7 days with 0.5 mM Fe(II)_(aq). Time 0 days represents conditions before Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ solids were added. Concentration units for (Fe) and (Mg) are mg/L. All data reported here have a standard deviation (RSD) of ±10%.

Time (Days)	MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
	Test 1			Test 1			Test 1			Test 1		
	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	9.0	0.07	41.6	9.7	0.07	1.08	9.4	0.11	90.0	9.2	0.09	5.05
2	9.0	0.5	59.8	9.7	1.95	1.87	9.2	0.21	100.4	9.2	0.05	5.48
4	9.1	0.34	46.1	9.7	0.14	0.79	9.0	0.21	101.9	9.2	0.06	3.99
7	9.1	0.43	38.4	9.8	0.09	0.66	9.9	0.10	115.9	9.0	0.50	2.30

Time (Days)	MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
	Test 2			Test 2			Test 2			Test 2		
	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	9.0	0.11	56.7	9.7	0.06	1.06	9.1	0.14	84.8	9.0	0.08	4.81
2	9.0	0.17	46.8	9.7	0.17	1.20	9.2	0.13	92.3	9.2	0.08	5.24
4	9.1	0.1	43.8	9.8	0.25	0.27	9.1	0.04	94.4	9.1	0.05	4.15
7	9.0	0.10	19.0	9.7	0.16	0.69	9.1	0.21	100. 7	9.0	0.17	4.55

Table S6. Leachability of Fe and Mg for individual tests conducted at pH 10 for 7 days with 0.5 mM Fe(II)_(aq). Time 0 days represents conditions Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ were added. Concentration units for (Fe) and (Mg) are mg/L. All data reported here have a standard deviation (RSD) of ±10%.

Time (Days)	MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
	Test 1			Test 1			Test 1			Test 1		
	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	9.2	1.03	20.1	10.1	0	0.60	9.2	0.10	53.1	9.8	0.20	0.64
2	9.3	0.07	10.4	9.9	0.08	0.53	9.3	0.10	69.4	10.1	0.1	0.40
4	9.7	0.07	3.90	9.9	0.13	0.43	9.4	0.19	58.9	9.9	0.18	0.34
7	9.7	0.21	2.31	9.9	0.15	0.46	9.4	0.04	39.0	9.9	0.30	0.57

Time (Days)	MgFeCO ₃			MgAlCO ₃			MgFeSO ₄			MgAlSO ₄		
	Test 2			Test 2			Test 2			Test 2		
	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)	pH	(Fe)	(Mg)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	9.2	0	17.4	9.9	0.11	0.74	9.3	0.2	53.9	9.7	0.08	0.60
2	9.3	0.07	9.70	9.9	0	0.45	9.4	0.09	64.8	10. 1	0.42	0.32
4	9.6	0.14	5.35	10. 0	0.09	0.44	9.6	0.16	45.8	10. 0	0.15	0.33
7	9.7	0.16	3.60	9.9	0.06	0.40	9.5	0.07	38.7	9.9	0.13	0.40

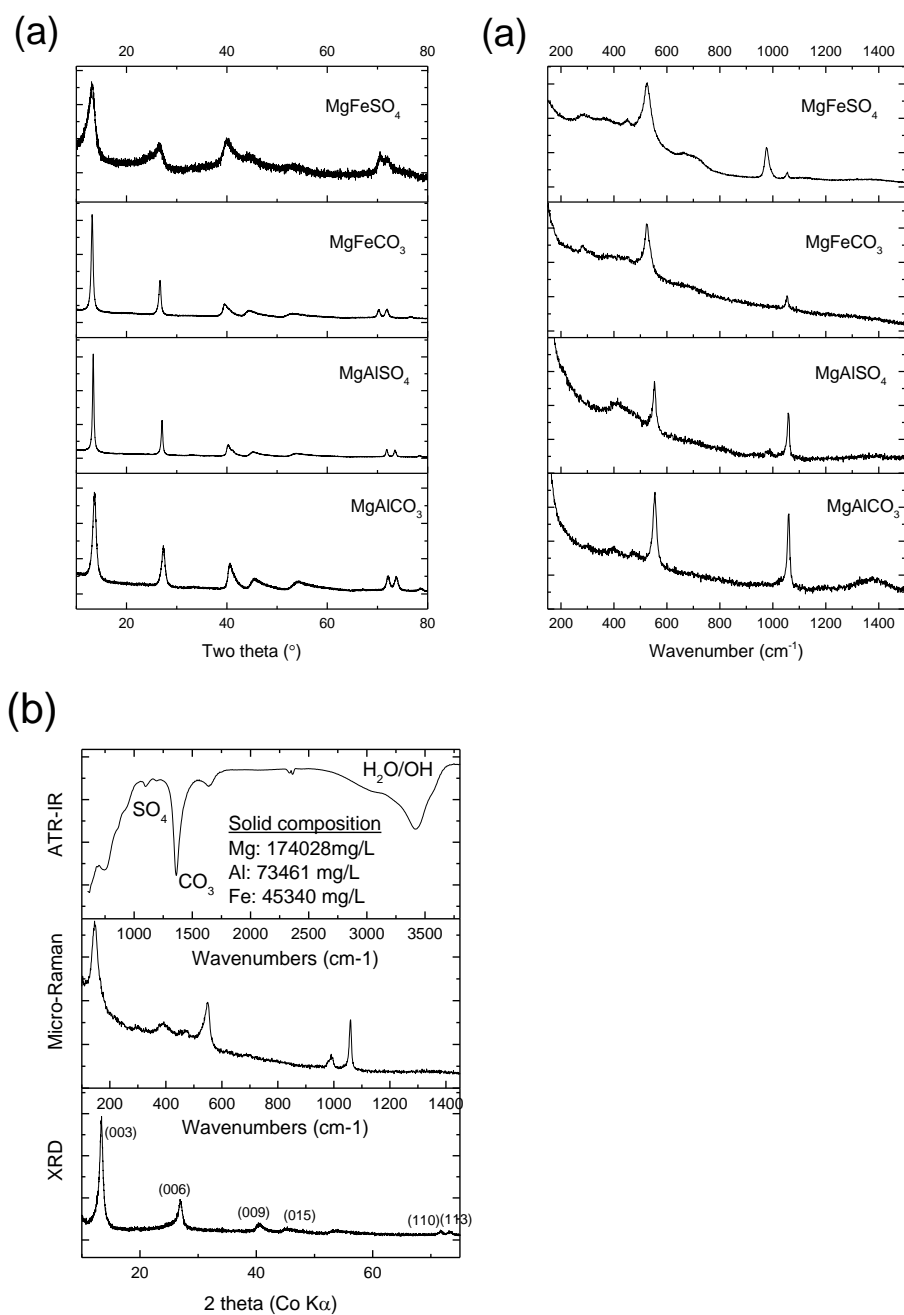


Figure S1. (a) XRD spectra (left) and micro-Raman spectra (right) of synthesized HTLC solid products before undergoing abiotic reduction. More detailed information on these starting materials can be found in our previous work.⁹ (b) ATR-IR, micro-Raman, and XRD spectra of the Mg(II)-Al(III)-Fe(III) containing- SO_4 - CO_3 HTLC synthetic standard used in our work shows the typical HTLC structure from the described methods. The elemental composition of this Mg(II)-Al(III)-Fe(III) containing- SO_4 - CO_3 HTLC solid is also included.

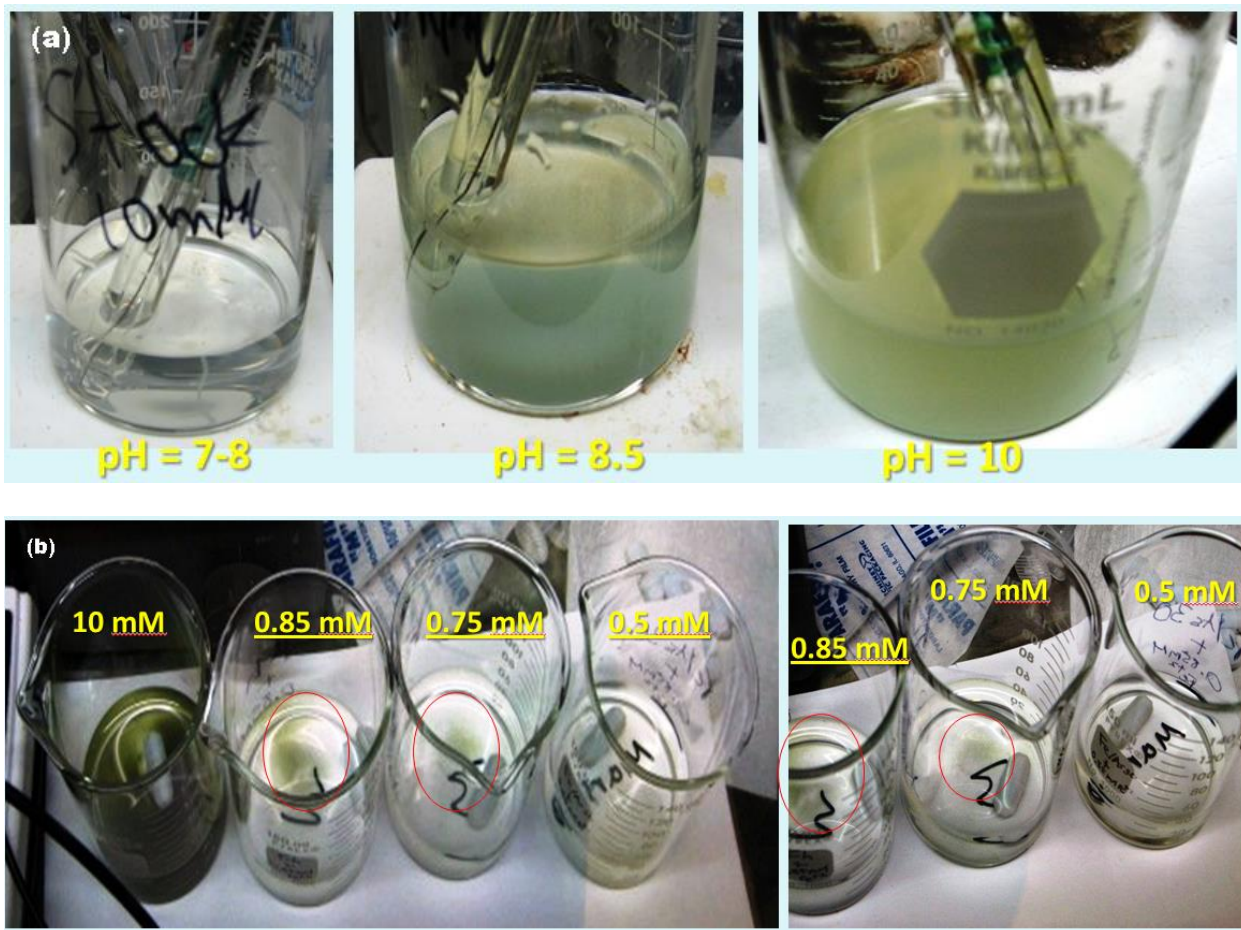


Figure S2. Precipitation of $\text{Fe(II)}_{(\text{aq})}$ in solution at (a) pH 7-10 and a concentration of 10 mM and (b) pH 10 at concentrations ranging from 0.5 to 10 mM.

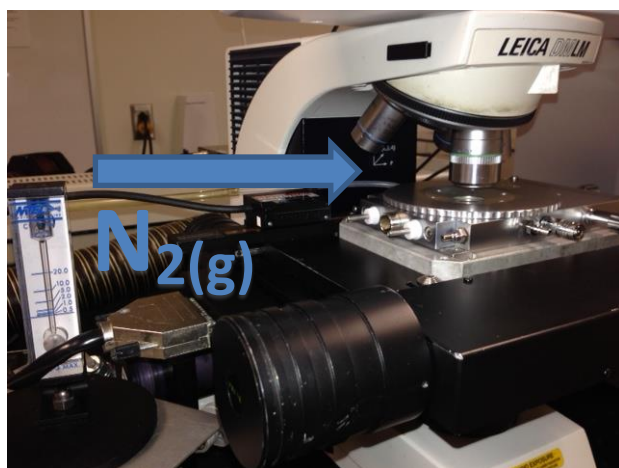


Fig S3. Inert (a) micro-Raman and (b) synchrotron based soft X-ray absorption spectroscopy experimental sample analysis set up.

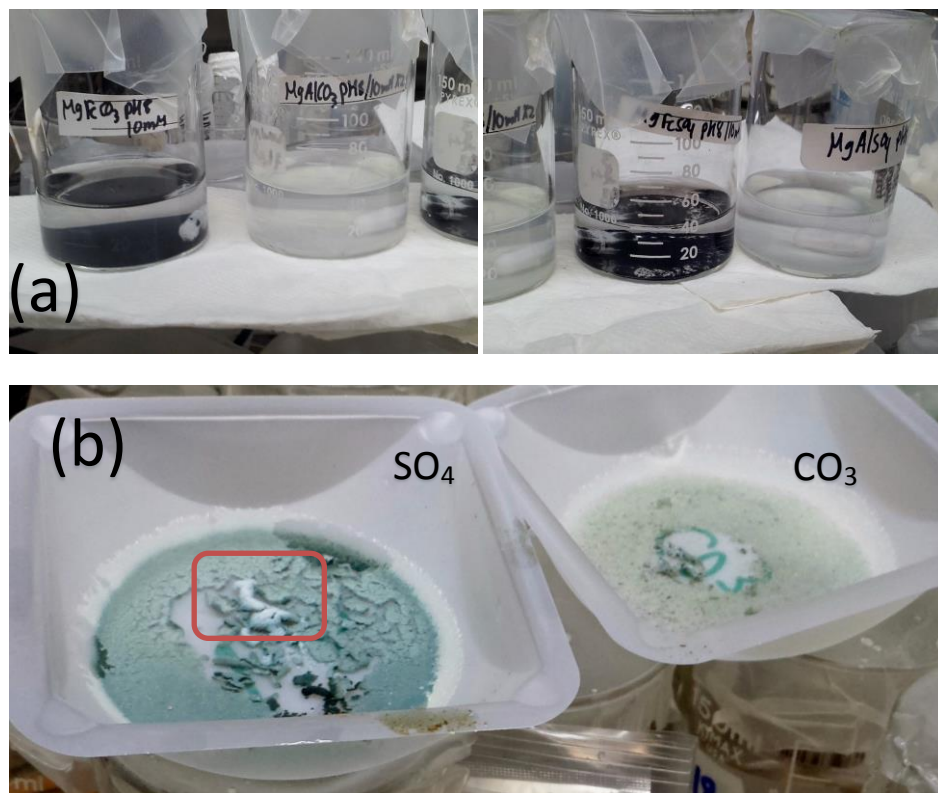


Fig S4. (a) Reacted Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ HTLCs at a target pH of 8 for 7 days and (b) reacted Mg-Al-CO₃/SO₄ HTLCs at a target pH of 8 for 1 day with 10 mM Fe(II)_(aq) compared to synthetic. The red box on the reacted Mg-Al-SO₄ HTLC shows a typical surface reaction precipitation.

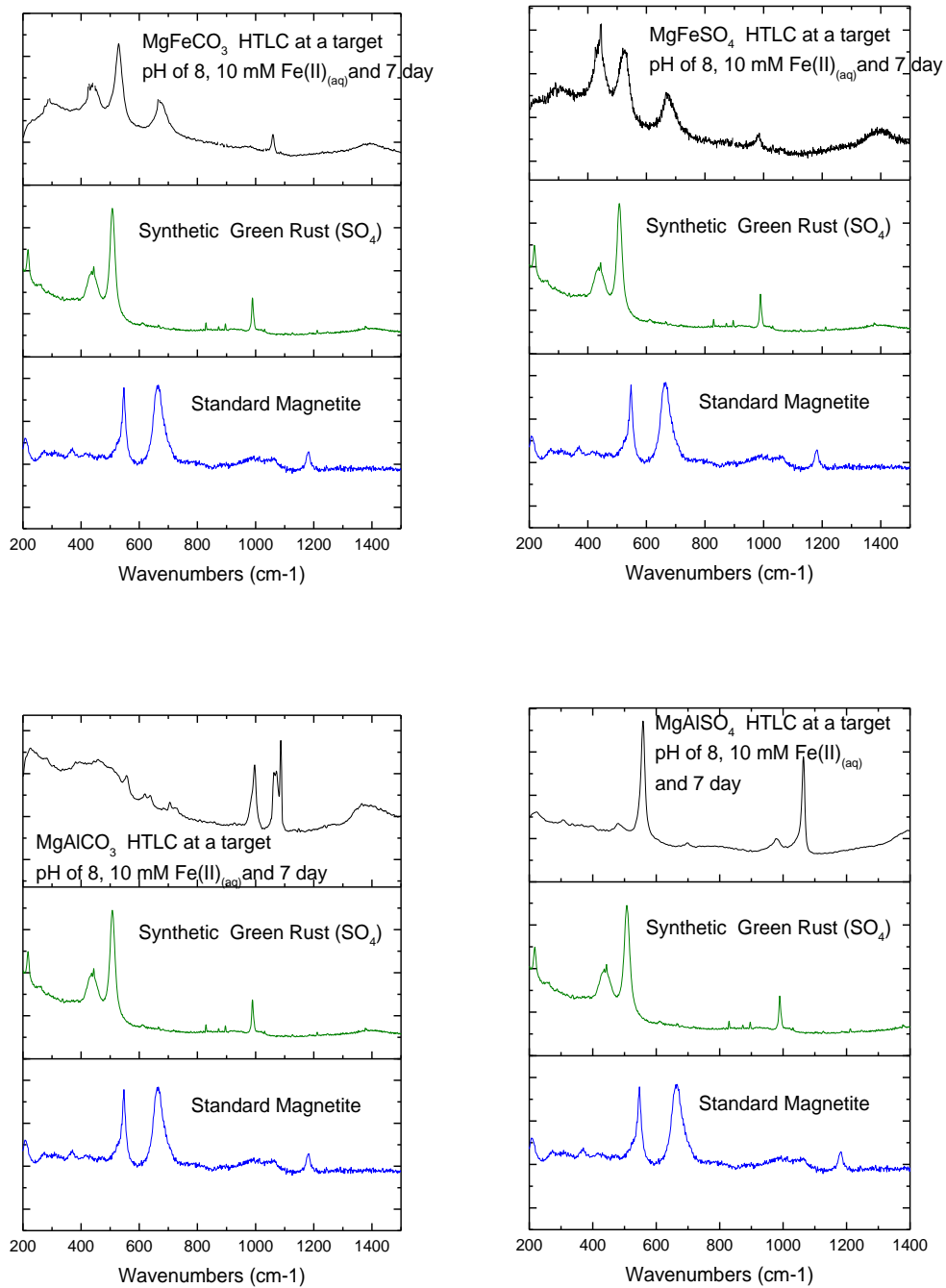


Fig S5. Inert micro-Raman spectra of the Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ sample reacted at a target pH of 8 for 7 days with 10 mM Fe(II)_(aq) compared to the standard GR(SO₄) and MG.

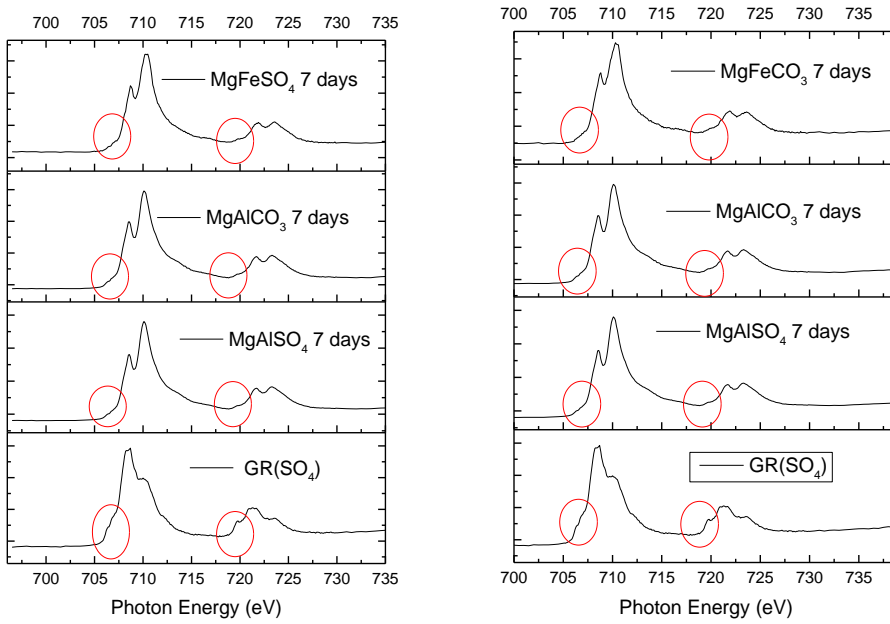
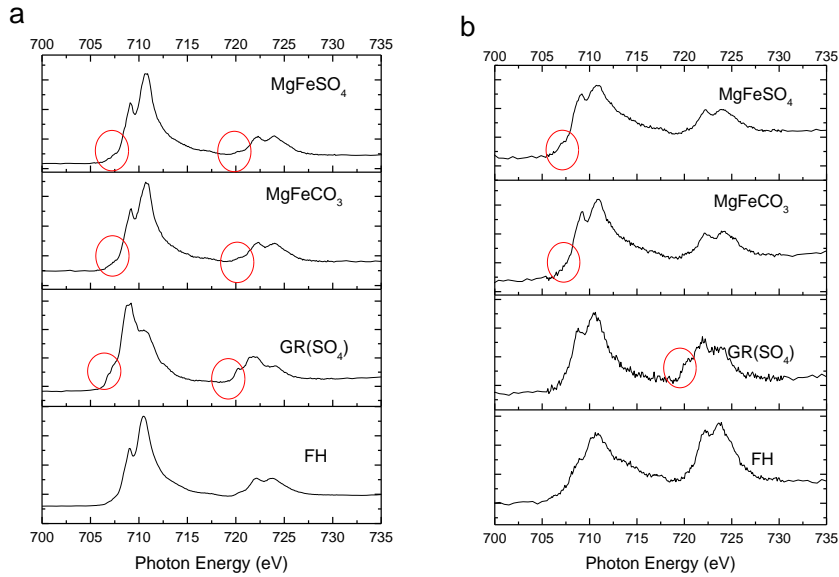


Fig S6. Fe L-edge TEY (surface) Mg(II)-Al(III)/Fe(III)-CO₃/SO₄ HTLCs at a target pH of 8, 10mM Fe(II)_(aq) for 7 days compared to standard GR. The red circles highlight the Fe(II) character of the spectra for the respective phases presented above. The energy scale presented in these spectra is relative.



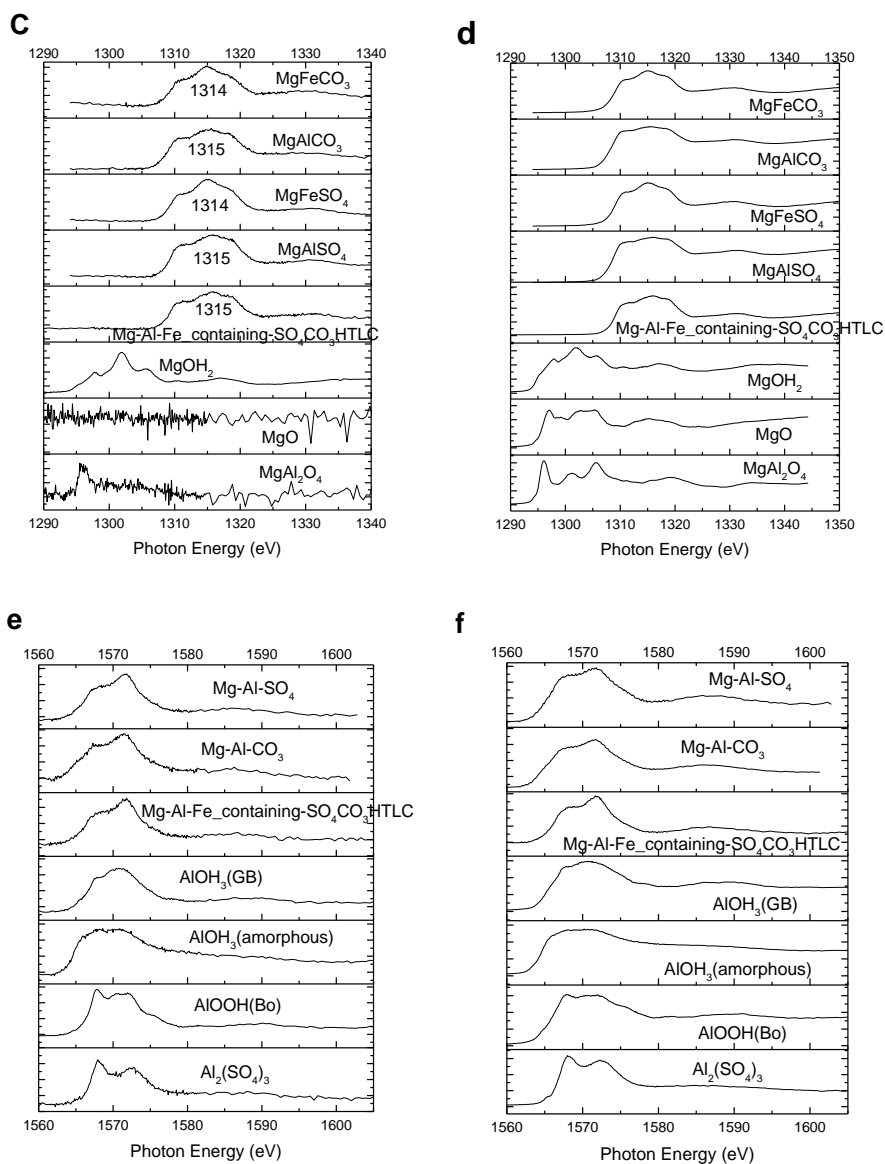


Fig S7. Energy calibrated Fe L-edge, Mg K-edge and Al K-edge TEY (surface: a, c and e) and PFY (bulk: b, d, f) spectra of reacted $\text{Mg(II)-Al(III)/Fe(III)-CO}_3/\text{SO}_4$ HTLCs at a target pH of 8, 10mM $\text{Fe(II)}_{(\text{aq})}$ for 7 days compared with relevant standards.