

Table S0. Initial composition of solids used in abiotic reduction tests. All units are in moles. Experimental ratio is the molar ratio of Fe/X.

Ferrihydrite			Fe/As 4			Fe/As 30			Fe/Mo 4			Fe/Mo 30			Fe/Ni 4			Fe/Ni 30		
Ideal			Ideal			Ideal			Ideal			Ideal			Ideal					
(Fe)	(X)	Exp Ratio	(Fe)	(X)	Exp Ratio	(Fe)	(X)	Exp Ratio	(Fe)	(X)	Exp Ratio	(Fe)	(X)	Exp Ratio	(Fe)	(X)	Exp Ratio	(Fe)	(X)	Exp Ratio
11.2	0	N/A	7.08	1.58	4.47	7.97	0.24	32.9	8.30	0.66	12.4	9.54	0.30	31.5	10.1	0.68	14.9	10.8	0.18	61.5

Table S1. The average and standard deviation of the pH range, % Fe removed from solution, and %X remaining in the solids after 7 days for tests conducted at pH 8 for 7 days with 10mM Fe(II)_(aq).

	Ferrihydrite	Fe/As 4.47	Fe/As 32.9	Fe/Mo 12.4	Fe/Mo 31.2	Fe/Ni 14.9	Fe/Ni 61.5
	Test 1	Test 1	Test 1	Test 1	Test 1	Test 1	Test 1
pH range measured	6.5-7.1	5.4-7.2	7.0-7.3	7.3-7.6	7.2-7.4	6.7-7.2	7.2-7.5
% Fe removed	81	98	88	91	78	98	99
%As, Mo, Ni remaining in solid	N / A	99	100	99	99	99	99

Table S2. The average and standard deviation of the pH range, % Fe removed from solution, and %X remaining in the solids after 7 days for tests conducted at pH 8 with 0.5mM Fe(II)_(aq).

	Ferrihydrite	Fe/As 4.47	Fe/As 32.9	Fe/Mo 12.4	Fe/Mo 31.2	Fe/Ni 14.9	Fe/Ni 61.5
	Test 1	Test 1	Test 1	Test 1	Test 1	Test 1	Test 1
pH range	6.7-7.1	5.6-6.1	6.7-7.2	7.9-8.2	7.7-8.0	7.2-7.9	7.3-7.6
% Fe removed	99	83	96	99	99	98	87
%As, Mo, Ni remaining in solid	N/A	99	100	99	99	99	99

Table S3. The average and standard deviation of the pH range, % Fe removed from solution, and %X remaining in the solids after 7 days for tests conducted at pH 10 with 0.5mM Fe(II)_(aq).

	Ferrihydrite	Fe/As 4.47	Fe/As 32.9	Fe/Mo 12.4	Fe/Mo 31.2	Fe/Ni 14.9	Fe/Ni 61.5
	Test 1	Test 1	Test 1	Test 1	Test 1	Test 1	Test 1
pH range	7.5-8.1	5.4-7.6	7.2-8.0	8.6-8.7	7.7-8.8	7.4-8.2	7.8-8.0
% Fe removed	99	99	99	99	99	97	99
%As, Mo, Ni remaining in solid	N/A	99	100	99	99	99	99

Table S4. Leachability of Fe and elements of concern (X = As, Mo, and Ni) for each individual test conducted at pH 8 for 7 days with 10mM Fe(II)_(aq). Time 0 days represents conditions before Fe/X solids were added. Concentration units for (Fe) and (X) are in ppm. All data reported here has standard deviation (RSD) of ±10%.

Time (Days)	Ferrihydrite			Fe/As 4.47			Fe/As 32.9			Fe/Mo 12.4			Fe/Mo 31.2			Fe/Ni 14.9			Fe/Ni 61.5		
	Test 1			Test 1			Test 1			Test 1			Test 1			Test 1			Test 1		
	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)
0	5.5	749	0	5.5	749	0	5.5	749	0	5.5	749	0	5.5	749	0	5.5	749	0	5.5	749	0
1	6.6	440	0	5.3	278	0.56	7.5	328	0	7.3	218	27.9	7.3	391	0.13	6.6	34.7	6.97	7.1	31.9	1.25
2	6.6	421	0	7.0	133	0.21	7.2	313	0	7.5	170	12.9	7.4	365	0.07	7.1	23.9	4.11	7.5	15.2	0.91
4	7.2	323	0	6.3	27.8	0.17	7.0	245	0	7.5	125	6.20	7.0	273	0.02	7.1	15.6	2.78	7.1	11.1	0.90
7	6.6	100	0	6.8	10.5	0.56	7.2	96.4	0	7.2	106	2.49	6.7	253	0.01	7.2	7.98	1.01	7.6	2.76	0.29
Time (Days)	Ferrihydrite			Fe/As 4.47			Fe/As 32.9			Fe/Mo 12.4			Fe/Mo 31.2			Fe/Ni 14.9			Fe/Ni 61.5		
	Test 2			Test 2			Test 2			Test 2			Test 2			Test 2			Test 2		
	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)
0	5.6	749	0	5.6	749	0	5.6	749	0	5.6	749	0	5.6	749	0	5.6	749	0	5.6	749	0
1	6.5	409	0	5.6	291	0.20	6.8	442	0	7.8	175	23.2	7.1	345	0.5	6.8	34.7	8.16	7.6	31.8	1.12
2	6.4	404	0	6.7	156	0.27	6.9	259	0	7.1	121	18.6	7.4	313	0.04	7.2	33.8	3.87	7.6	29.2	1.08

4	7.1	316	0	7.3	34.	0.50	7.6	170	0	7.7	69.0	11.9	7.6	69.0	0.01	7.1	11.4	1.49	7.4	13.5	0.95
7	6.4	181	0	7.6	12.3	0.30	7.3	79	0	7.7	29.1	1.20	7.5	68.0	0.01	7.3	8.02	1.20	7.5	5.95	0.39

Table S5. Leachability of Fe and elements of concern (X = As, Mo, and Ni) for each individual test conducted at pH 8 for 7 days with 0.5 mM Fe(II)_(aq). Time 0 days represents conditions before Fe/X solids were added. Concentration units for (Fe) and (X) are in ppm. All data reported here has standard deviation (RSD) of ±10%.

Time (Days)	Ferrihydrite			Fe/As 4.47			Fe/As 32.9			Fe/Mo 12.4			Fe/Mo 31.2			Fe/Ni 14.9			Fe/Ni 61.5		
	Test 1			Test 1			Test 1			Test 1			Test 1			Test 1			Test 1		
	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	7.2	6.18	0	5.7	4.9	15.8	6.5	9.41	0	7.9	0.03	220	7.8	0.12	3.25	7.24	20.8	181	7.3	16.8	27.7
2	6.7	3.56	0	5.9	1.56	17.8	7.1	7.68	0	8.1	0.03	260	7.8	0.05	12.7	7.6	1.68	52.8	7.4	7.89	14.4
4	6.6	0.92	0	6.1	0.57	29.4	7.0	5.04	0	8.0	0.02	308	8.2	0.01	15.9	7.8	1.42	38.9	7.6	5.71	14.1
7	6.9	0.27	0	6.2	0.26	29.8	6.5	1.42	0	8.1	0.02	340	7.6	0.01	16.1	8.1	0.73	14.6	7.7	3.33	8.68
Time (Days)	Ferrihydrite			Fe/As 4.47			Fe/As 32.9			Fe/Mo 12.4			Fe/Mo 31.2			Fe/Ni 14.9			Fe/Ni 61.5		
	Test 2			Test 2			Test 2			Test 2			Test 2			Test 2			Test 2		
	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	6.3	7.43	0	5.6	9.4	10.8	7.3	14.6	0	7.9	0.04	275	7.8	7.68	3.61	7.2	14.2	136	7.3	31.9	18.7

2	7.5	4.61	0	5.5	2.8	17.8	7.3	2.6	0	8.3	0.04	260	7.8	0.02	3.59	7.8	1.68	63.2	7.5	19.2	11.3
4	7.2	1.13	0	6.2	0.43	32.3	7.4	2.82	0	8.1	0.03	309	7.9	0.02	5.01	7.8	1.4	42.5	7.6	13.6	15.6
7	6.6	0.33	0	6.1	0.10	32.6	6.9	1.27	0	7.9	0.03	341	7.8	0.02	6.37	7.81	0.7	10.7	7.5	5.95	12.5

Table S6. Leachability of Fe and elements of concern (X = As, Mo, and Ni) for each individual test conducted at pH 10 for 7 days with 0.5mM Fe(II)_(aq). Time 0 days represents conditions before Fe/X solids were added. Concentration units for (Fe) and (X) are in ppm. All data reported here has standard deviation (RSD) of ±10%.

Time (Days)	Ferrihydrite Test 1			Fe/As 4.47 Test 1			Fe/As 32.9 Test 1			Fe/Mo 12.4 Test 1			Fe/Mo 31.2 Test 1			Fe/Ni 14.9 Test 1			Fe/Ni 61.5 Test 1		
	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0
1	7.8	1.59	0	5.2	5.49	0.92	5.8	10.3	0	8.5	0.04	376	8.7	0.05	8.21	7.3	0.03	61.8	7.9	3.20	5.11
2	7.8	0.27	0	5.0	2.03	3.65	7.0	2.94	0	8.8	0.03	429	7.7	0.04	24.7	8.0	0.03	13.9	8.0	0.18	0.43
4	8.2	0.13	0	7.1	0.20	6.80	7.4	0.37	0	9.0	0.02	429	8.7	0.02	35.4	8.1	0.03	7.56	8.1	0.46	0.70
7	7.9	0.04	0	7.6	0.07	9.80	7.4	0.17	0	8.9	0.02	385	8.8	0.02	46.1	8.1	0.03	1.29	7.8	0.46	0.13
Time (Days)	Ferrihydrite Test 2			Fe/As 4.47 Test 2			Fe/As 32.9 Test 2			Fe/Mo 12.4 Test 2			Fe/Mo 31.2 Test 2			Fe/Ni 14.9 Test 2			Fe/Ni 61.5 Test 2		
	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)	pH	(Fe)	(X)
0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0	5.5	38	0

1	8.1	0.2	0	6.1	5.49	2.0	8.6	0.09	0	8.7	0.03	376	7.9	0.05	3.5	7.6	86.2	86.2	7.8	2.2	5.5
2	7.2	0.05	0	5.9	2.0	1.9	8.3	0.09	0	8.4	0.02	321	8.6	0.04	8.2	8.1	7.64	7.64	8.1	2.2	0.97
4	8.1	0.03	0	7.6	0.2	6.8	8.7	0.08	0	8.5	0.01	470	8.5	0.03	35.3	8.2	4.7	4.7	7.9	0.6	0.91
7	8.0	0.03	0	7.7	0.07	10.5	8.5	0.07	0	8.6	0.01	490	8.6	0.03	46.1	8.3	2.11	2.11	8.0	0.3	0.06

Table S7. Phase transformation to lepidocrocite (LP), goethite (GT), green rust-SO₄ (GR), or magnetite (MG) based on results from XRD and micro-Raman spectroscopy for tests conducted at pH 8 for 7 days and with 10mM Fe(II)_(aq).

Sample	No phase transformation			Phase transformation [‡]		
	XRD	Raman	TEM [‡]	XRD	Raman	TEM [‡]
Ferrihydrite	-	-	-	LP+GT	LP+GT	-
Fe/As 4.47	X	X	X	-	-	-
Fe/As 32.9	-	-	-	LP+GT	LP+GT	-
Fe/Mo 12.4	-	-	-	GT	GT	Yes
Fe/Mo 31.5	-	-	-	LP+GT	LP+GT	Yes
Fe/Ni 14.9	-	-	-	GT+MG	GT+MG	-
Fe/Ni 61.5	-	-	-	LP+MG	LP+MG	-

[‡] Phase transformation was not observed to be 100% via XRD (see Fig. 1) but indications of its transformation were observed in the XRD and micro-Raman data. Determination of % phase transformation was out of the scope of this work.

[‡] TEM was only done on selected samples and was only used to confirm whether crystallization from the amorphous ferrihydrite matrix occurred.

Table S8. Phase transformation to lepidocrocite (LP), goethite (GT), magnetite (MG), or green rust-SO₄ (GR) based on results from XRD and micro-Raman spectroscopy (see supplementary information for data) for tests conducted at pH 8 for 7 days and with 0.5mM Fe(II)_(aq).

Sample	No phase transformation			Phase transformation [‡]		
	XRD	Raman	TEM [‡]	XRD	Raman	TEM [‡]
Ferrihydrite	-	-	-	LP+GT	LP+GT	-
Fe/As 4.47	X	X	X	-	-	-
Fe/As 32.9	-	-	-	LP	LP	-
Fe/Mo 12.4 [*]	X	X	-	-	-	Yes
Fe/Mo 31.5	-	-	-	-	LP+GT	Yes
Fe/Ni 14.9	-	-	-	-	GR	-
Fe/Ni 61.5	-	-	-	GT	LP+GT	-

[‡] Phase transformation was not observed to be 100% via XRD (see Fig. 1) but indications of its transformation were observed in the XRD and micro-Raman data. Determination of % phase transformation was out of the scope of this work.

[‡] TEM was only done on selected samples and was only used to confirm whether crystallization from the amorphous ferrihydrite matrix occurred.

^{*} TEM images (see below in Fig. S7) show the presence of a nano-crystalline phase that was too small to detect via XRD and micro-Raman spectroscopy.

Table S9. Phase transformation to lepidocrocite (LP), goethite (GT), magnetite (MG), or green rust-SO₄ (GR) based on results from XRD and micro-Raman spectroscopy for tests conducted at pH 10 for 7 days and with 0.5mM Fe(II)_(aq).

Sample	No phase transformation			Phase transformation [‡]		
	XRD	Raman	TEM [‡]	XRD	Raman	TEM [‡]
Ferrihydrite	-	-	-	-	LP	-
Fe/As 4.47	X	X	X	-	-	-
Fe/As 32.9	-	-	-	-	LP	-
Fe/Mo 12.4 [*]	X	X	X	-	-	-
Fe/Mo 31.5 [*]	X	X	X	-	-	-
Fe/Ni 14.9	-	-	-	-	GT+MG	-
Fe/Ni 61.5	-	-	-	-	MG	-

[‡] Phase transformation was not observed to be 100 % via XRD (see Fig. 1) but indications of its transformation were observed in the XRD and micro-Raman data. Determination of % phase transformation was out of the scope of this work.

[‡] TEM was only done on selected samples and was only used to confirm whether crystallization from the amorphous ferrihydrite matrix occurred.

^{*} TEM images (see below Fig. S10) do not show the presence of nano-crystalline phases such as LP and/or GT.

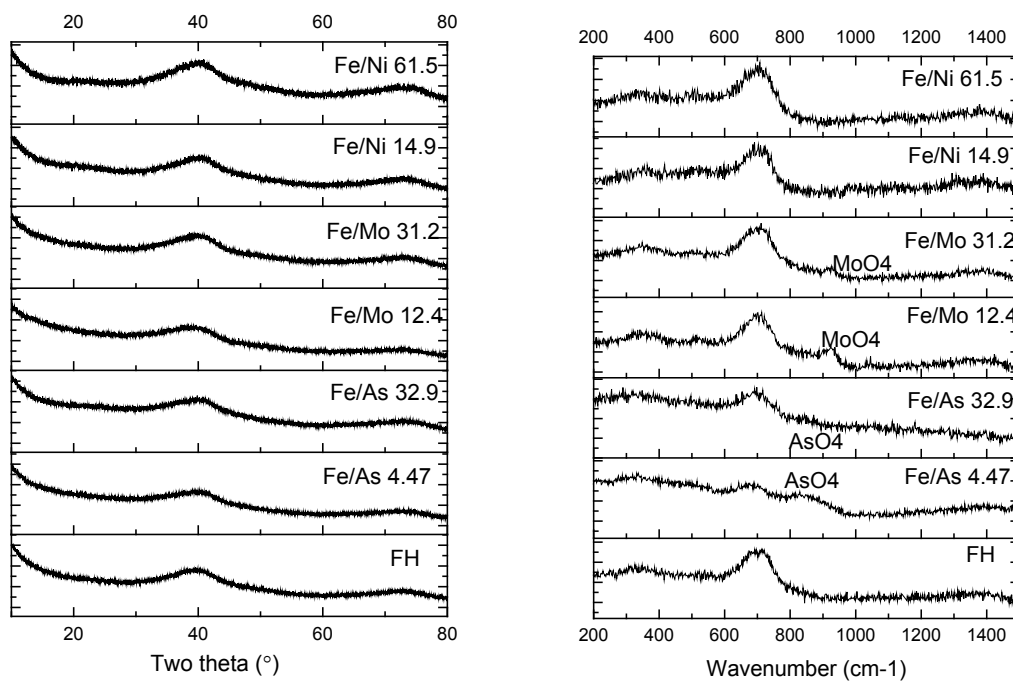


Figure S1. XRD spectra (left) and micro-Raman spectra (right) of synthesized solid products before undergoing abiotic reduction. The XRD and Raman show a typical 2-line ferrihydrite with the corresponding elements-molecules. In the case of Ni-FH, the Ni-O-Fe bond is not visible in the Raman spectra.

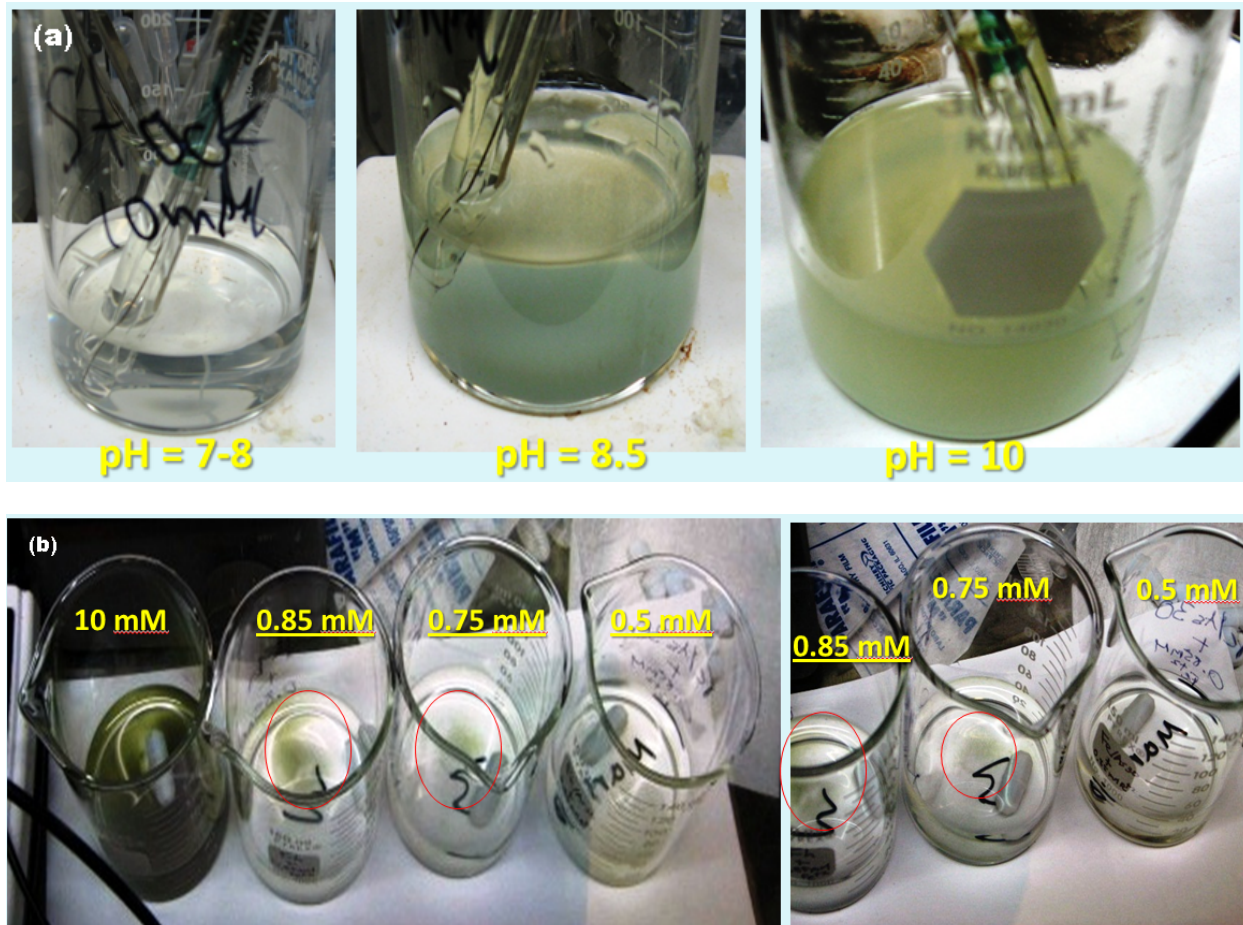


Figure S2. Precipitation of $\text{Fe(II)}_{(\text{aq})}$ in solution at (a) pH 7-10 and a concentration of 10mM and (b) pH 10 at concentrations ranging from 0.5 to 10mM.

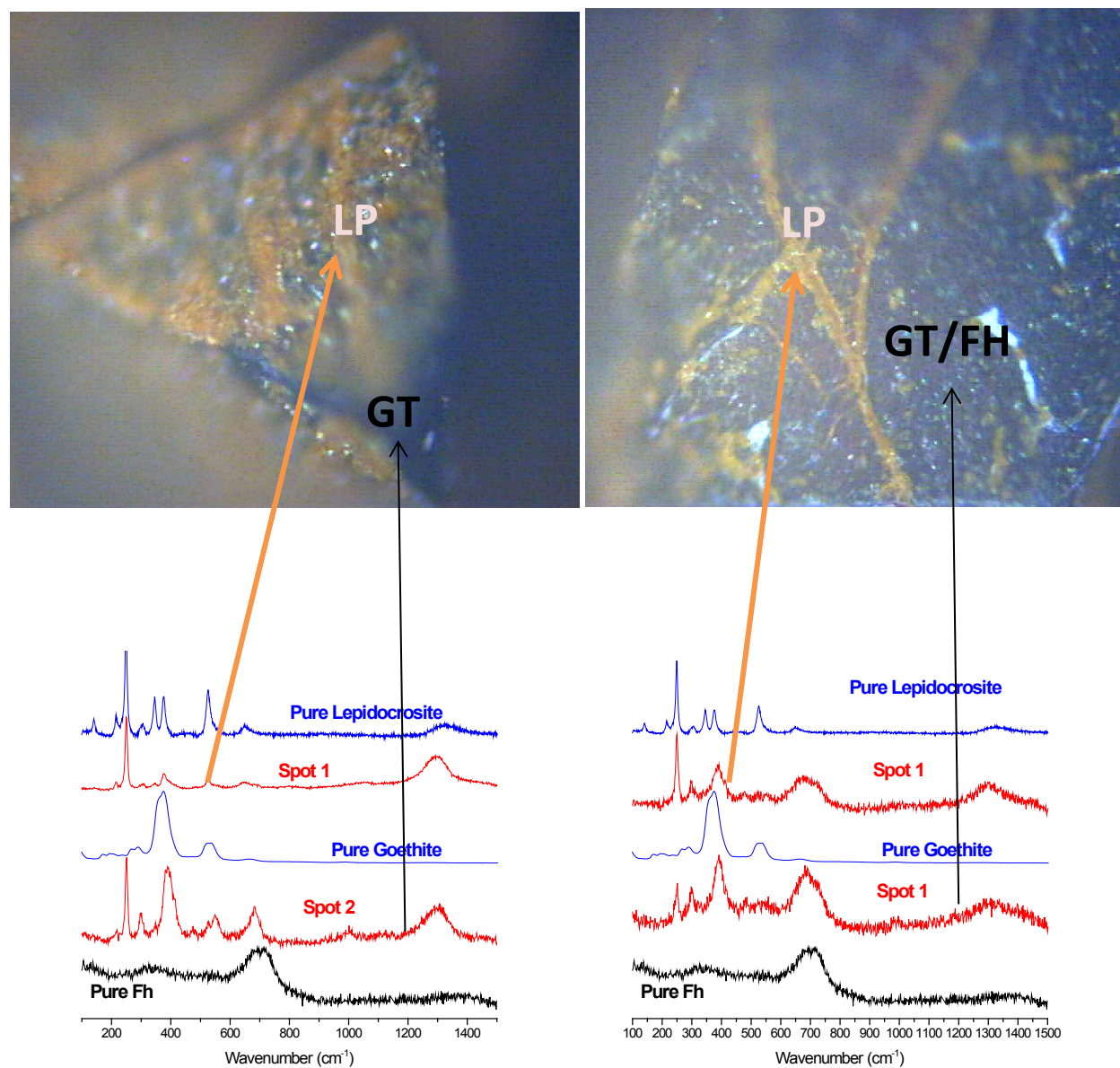
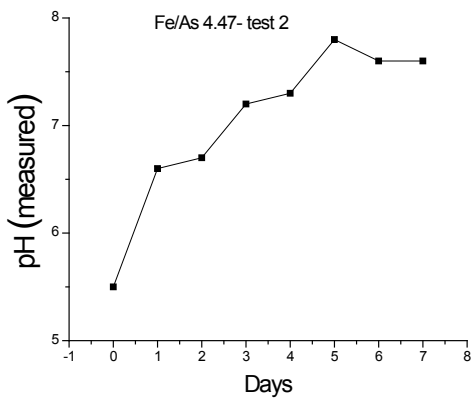
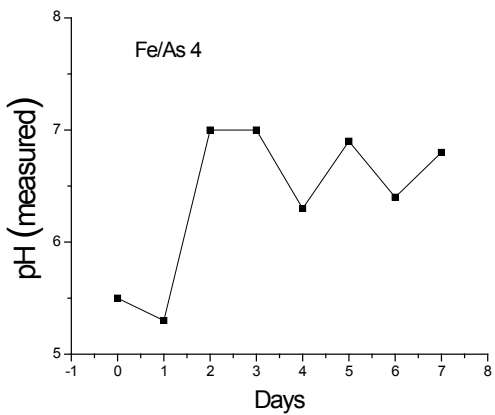
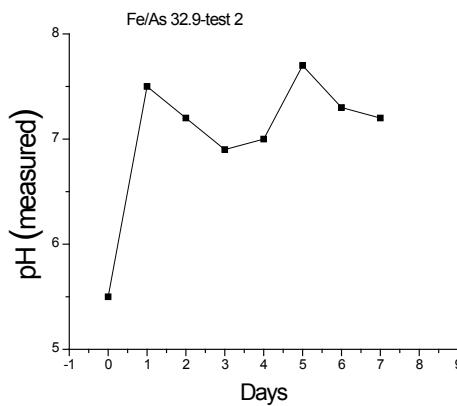
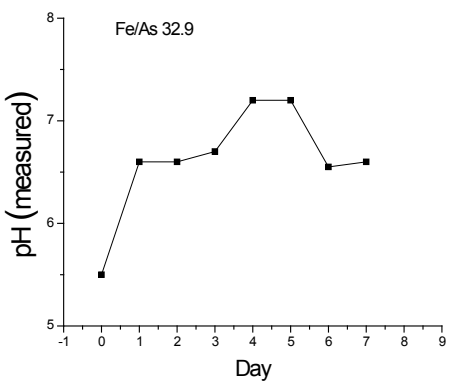
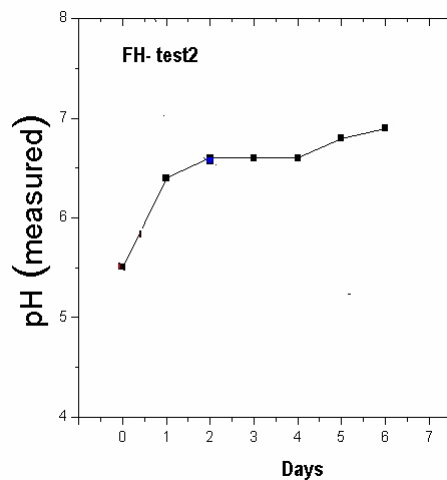
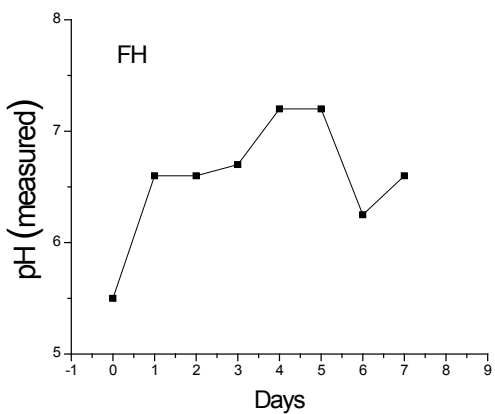
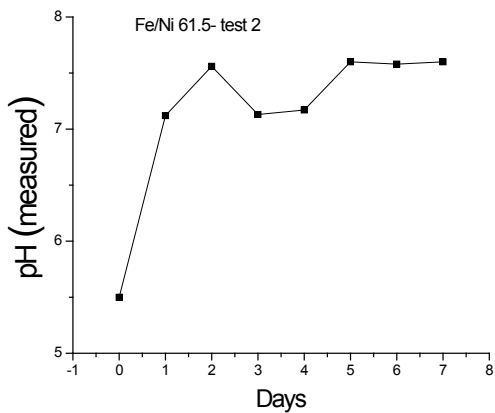
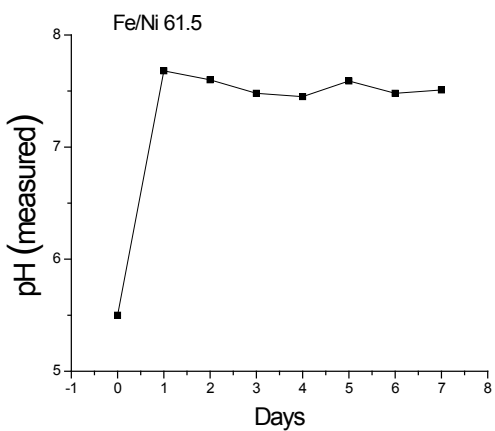
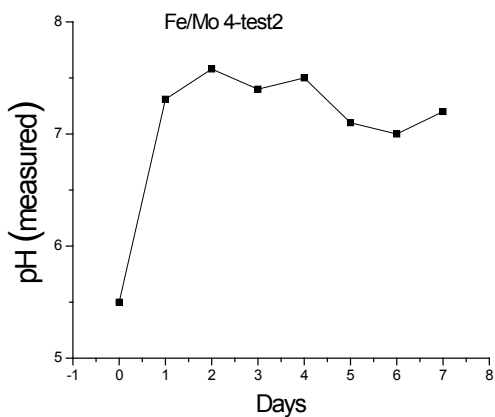
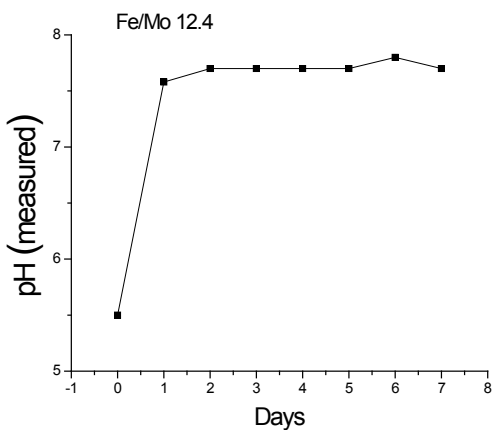
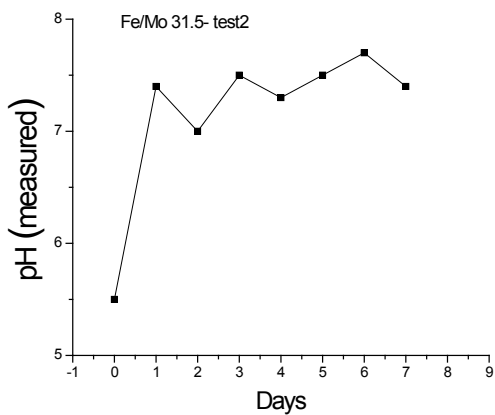
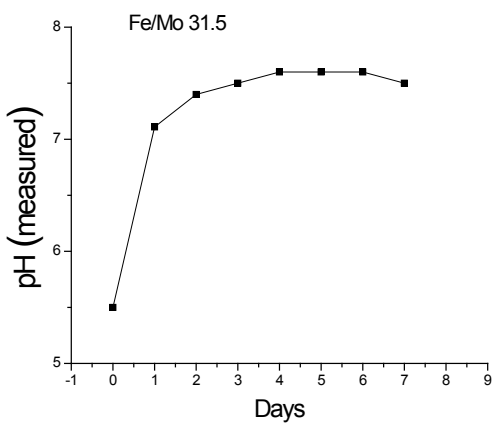


Figure S3. Light microscope images (using the Raman microscope) of FH particles reacted for 7 days at a target pH of 8 with 10mM (left) and 0.5mM (right) Fe(II)_(aq). The Raman spectra of the indicated spot are also shown below each figure and compared with LP, GT, and FH standards.





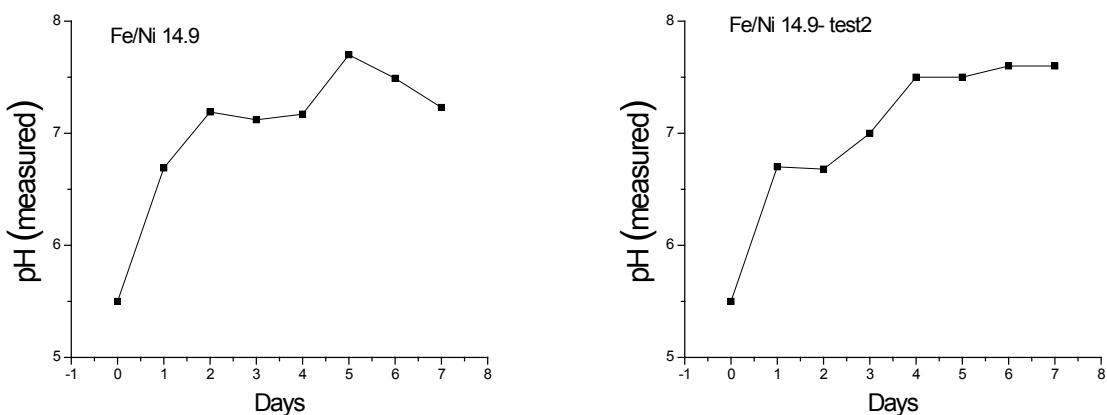


Fig S4. Measured pH profiles of FH, Fe/As 32.9, Fe/As 4.47, Fe/Mo 32.9, Fe/Mo 12.4, Fe/Ni 61.5, and Fe/Ni 14.9 reacted at a target pH of 8 for 7 days with 10mM Fe(II)_(aq). No error bars on the individual measurements were added but all have a standard deviation (RSD) of $\pm 10\%$.

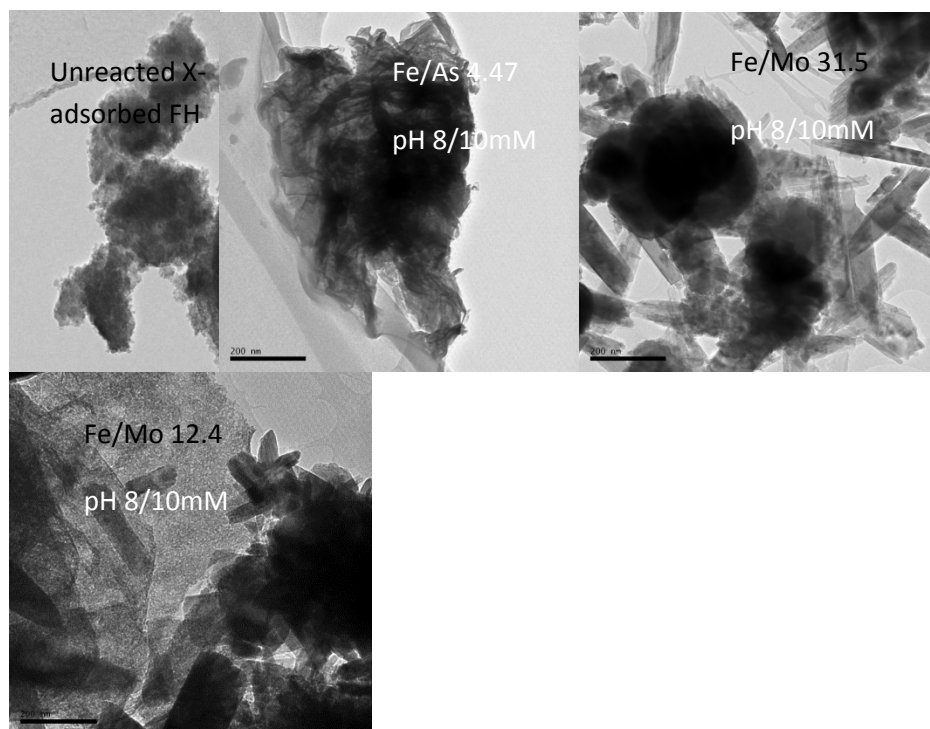
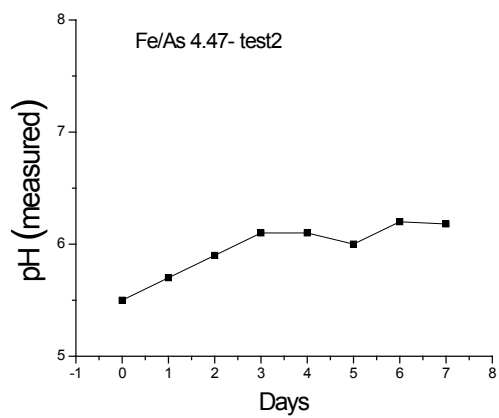
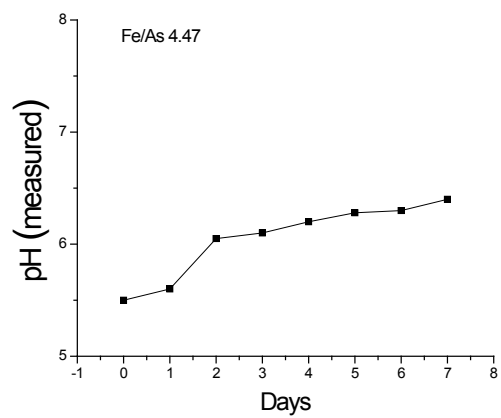
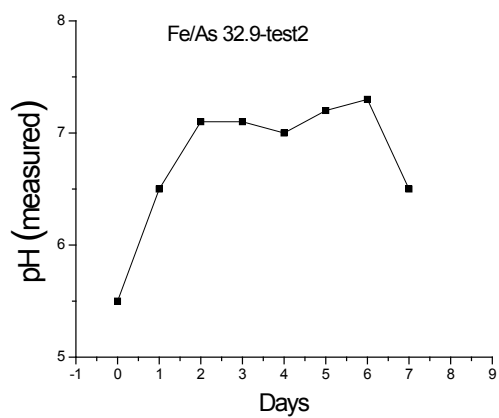
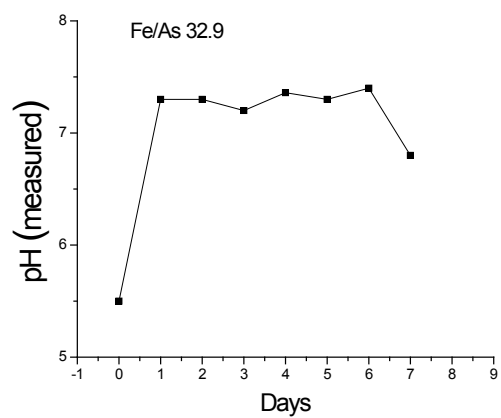
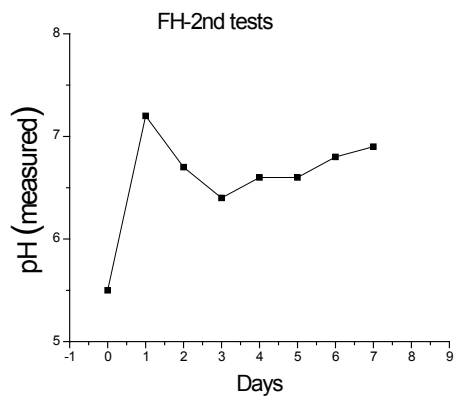
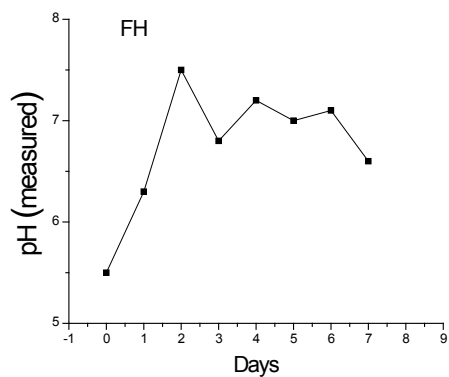
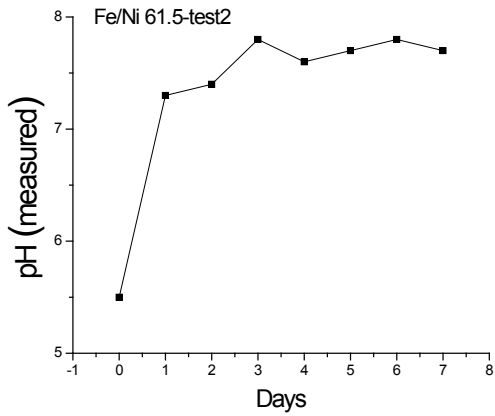
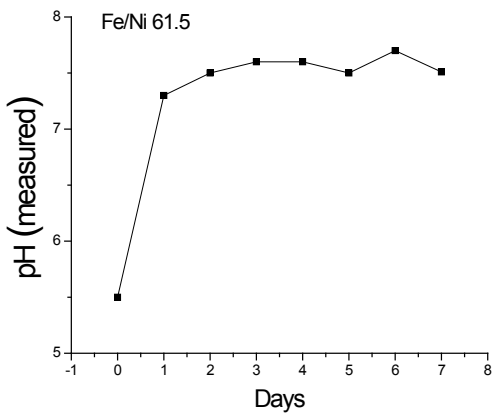
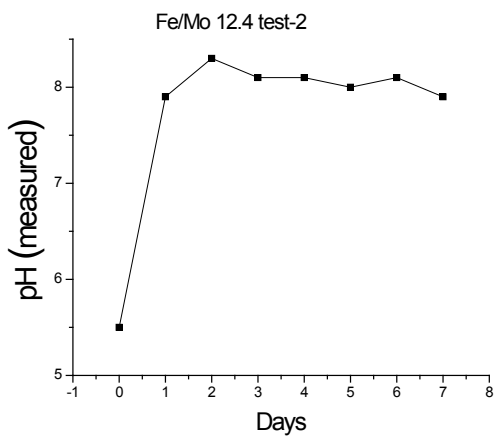
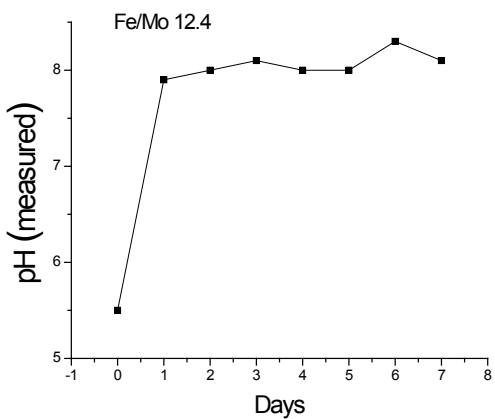
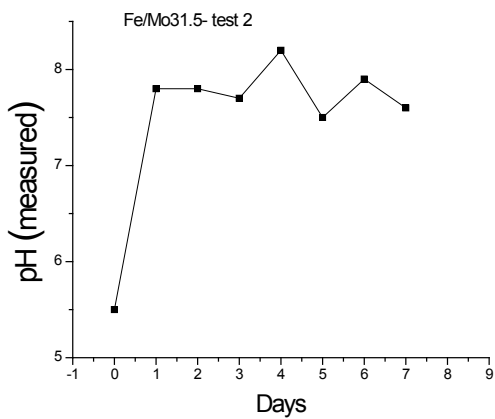
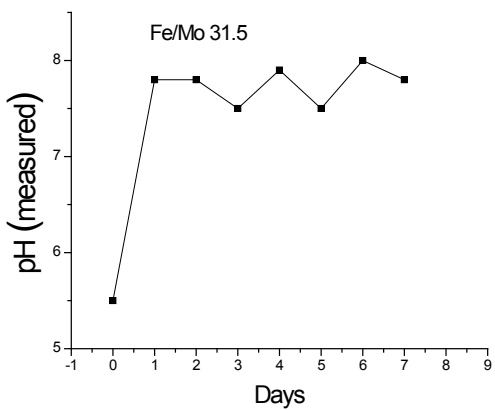


Fig S5. TEM of selected products after reaction for 7 days at a target pH of 8 and concentration of 10mM Fe(II)_(aq). The initial unreacted FH is amorphous aggregates and similar to that observed at Fe/As 4.47. In contrast, sharp and long needle-like particles in addition to the FH matrix are visible for both Fe/Mo reacted samples.





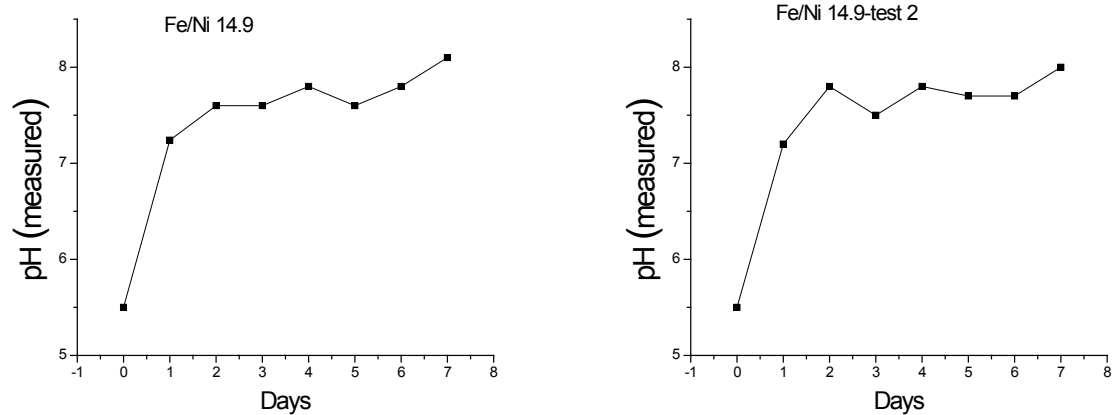


Fig S6. Measured pH profiles of FH, Fe/As 32.9, Fe/As 4.47, Fe/Mo 32.9, Fe/Mo 12.4, Fe/Ni 61.5, and Fe/Ni 14.9 reacted at a target pH of 8 for 7 days with 0.5mM Fe(II)_(aq). No error bars on the individual measurements were added but all have a standard deviation (RSD) of ±10%.

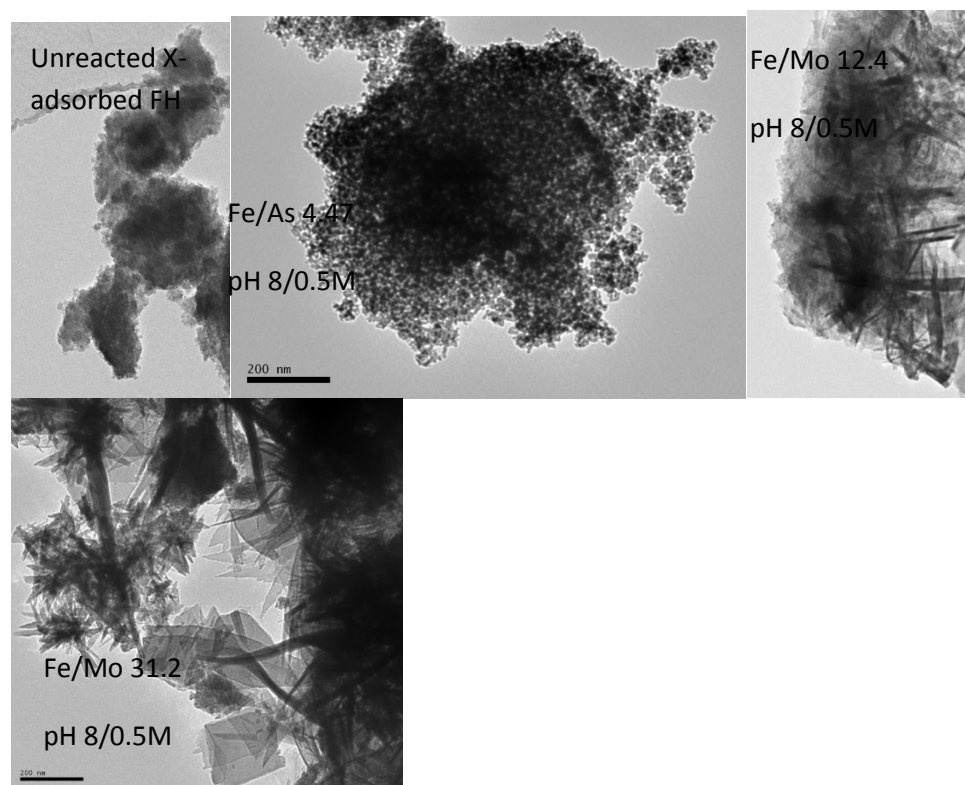


Fig S7. TEM of selected products after reaction for 7 days at a target pH of 8 and a concentration of 0.5mM $\text{Fe(II)}_{(\text{aq})}$. The initial unreacted FH is amorphous aggregates and similar to that observed at Fe/As 4.47. In contrast, sharp and long needle-like particles in addition to the FH matrix are visible for both Fe/Mo reacted samples.

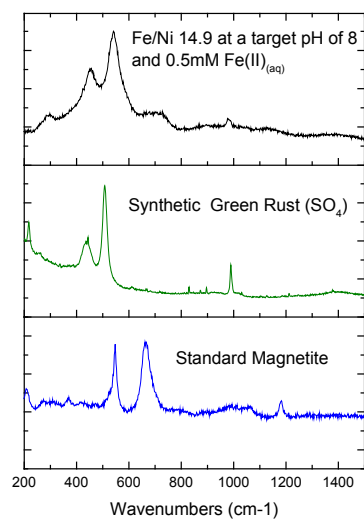
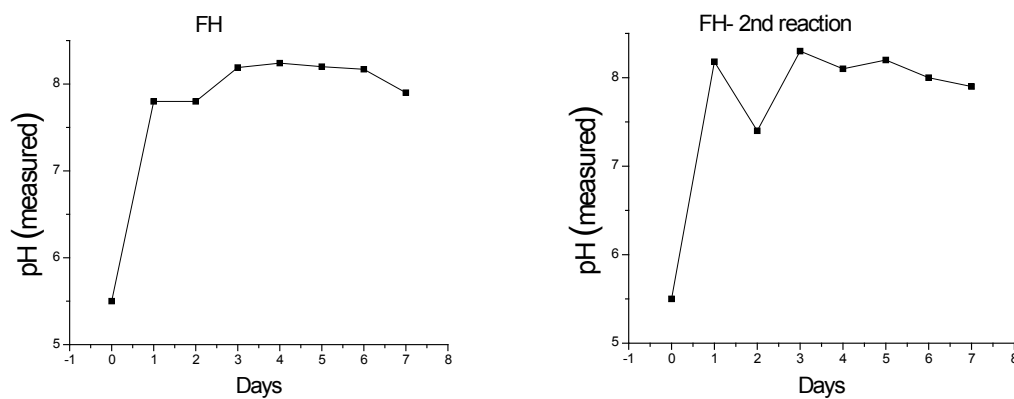
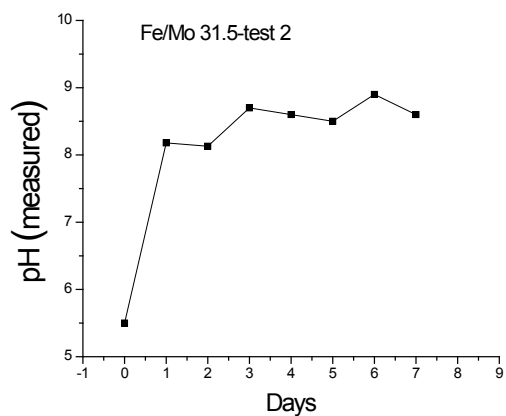
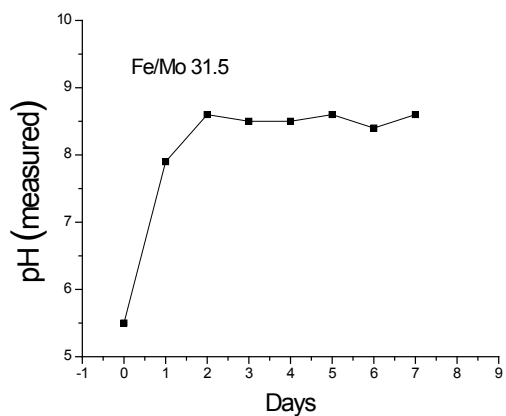
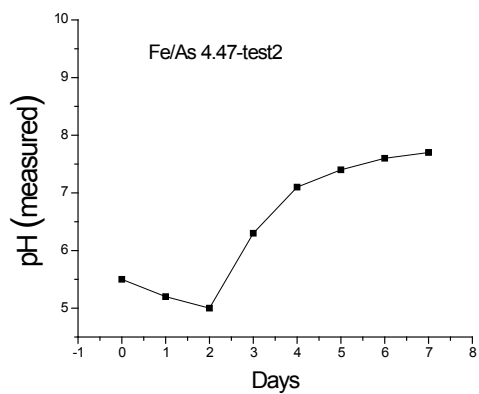
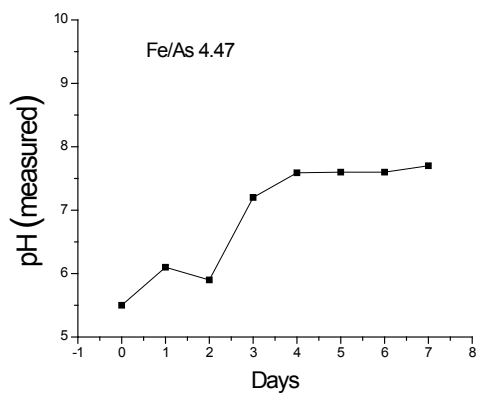
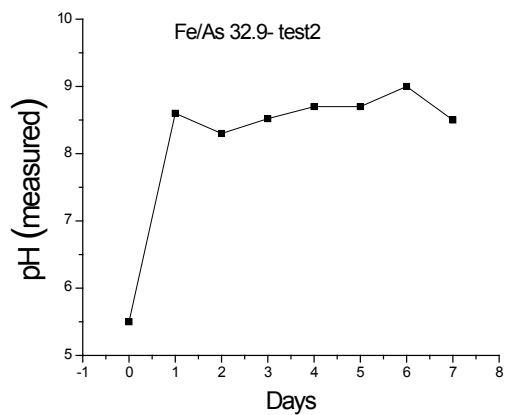
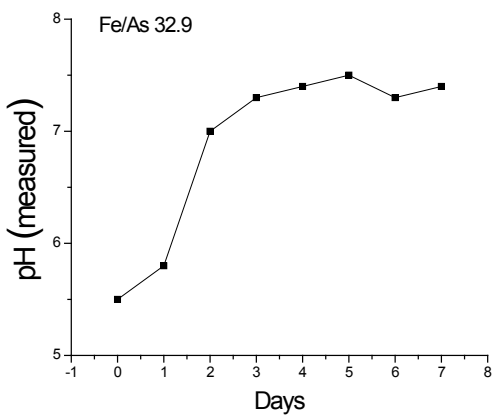


Fig S8. Raman spectra of the Fe/Ni 14.9 sample reacted at a target pH of 8 for 7 days with 0.5mM $\text{Fe(II)}_{(\text{aq})}$ compared to synthetic sulfated GR and reagent grade magnetite (Sigma-Aldrich).





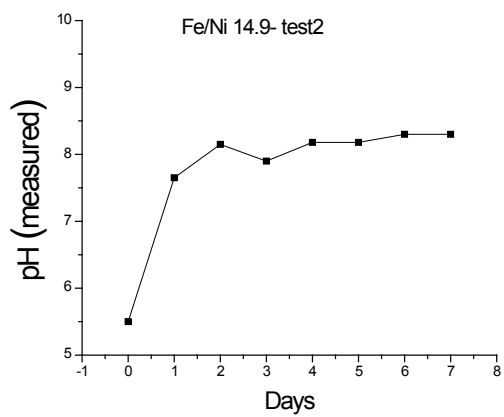
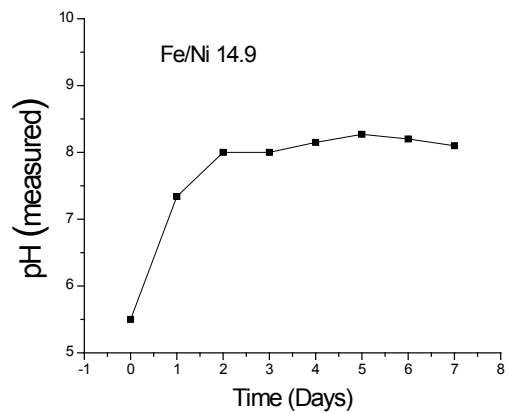
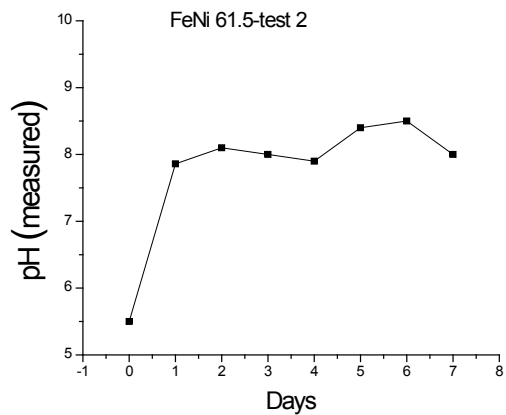
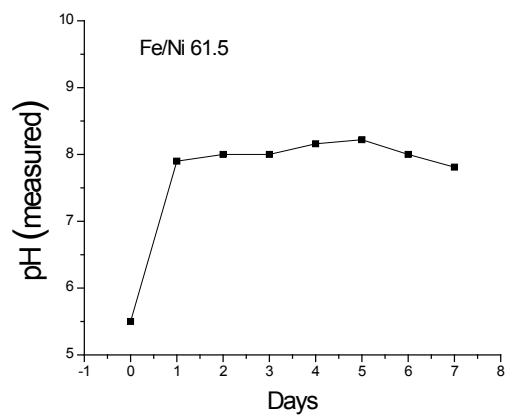
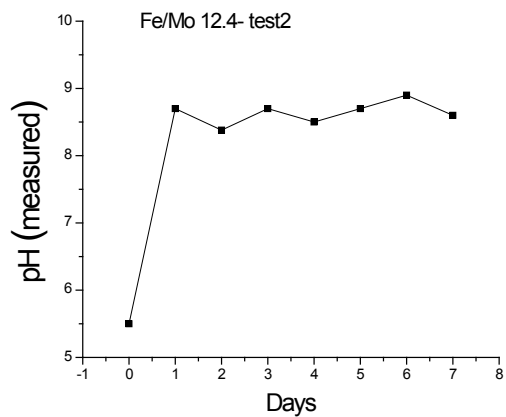
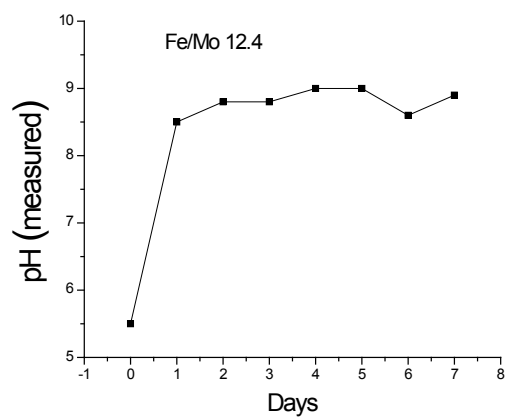


Fig S9. Measured pH profiles of FH, Fe/As 32.9, Fe/As 4.47, Fe/Mo 32.9, Fe/Mo 12.4, Fe/Ni 61.5, and Fe/Ni 14.9 reacted at a target pH of 10 for 7 days with 0.5mM Fe(II)_(aq). No error bars on the individual measurements were added but all have a standard deviation (RSD) of ±10%.

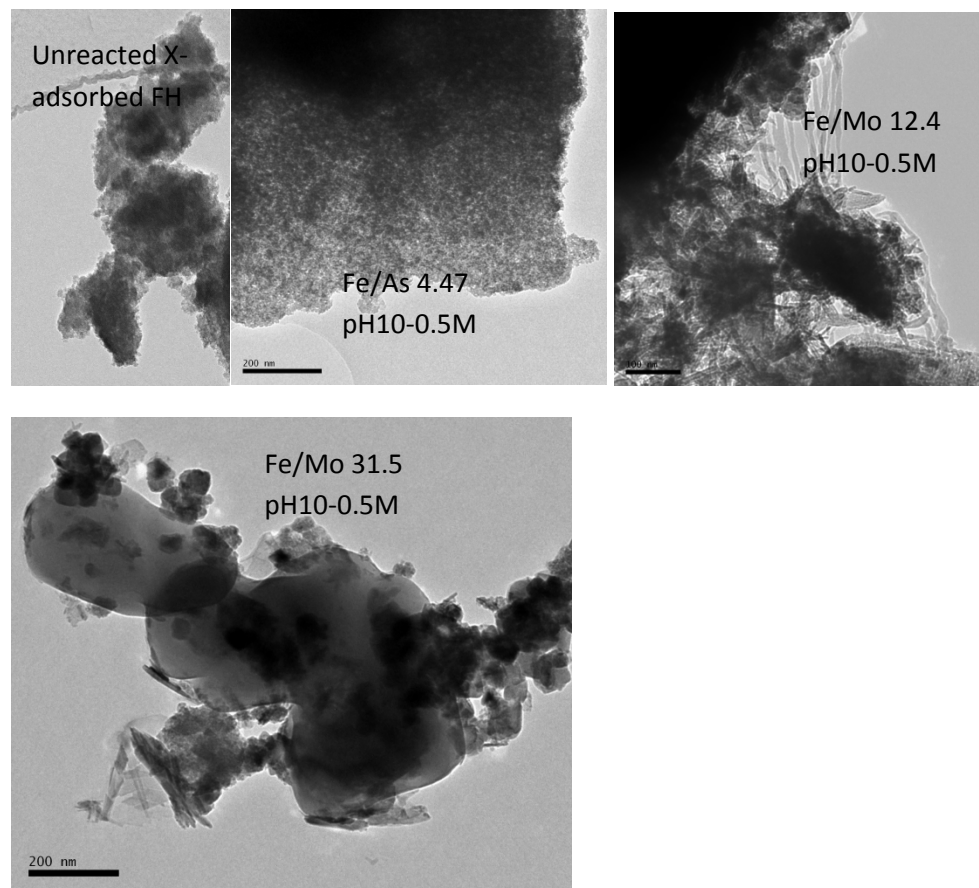


Fig S10. TEM of selected products after reaction for 7 days at a target pH of 10 and a concentration of 0.5mM Fe(II)_(aq). The initial unreacted FH is amorphous aggregates and similar to that observed at Fe/As 4.47. Interestingly, both Fe/Mo reacted samples show a lack of needle particle formation.

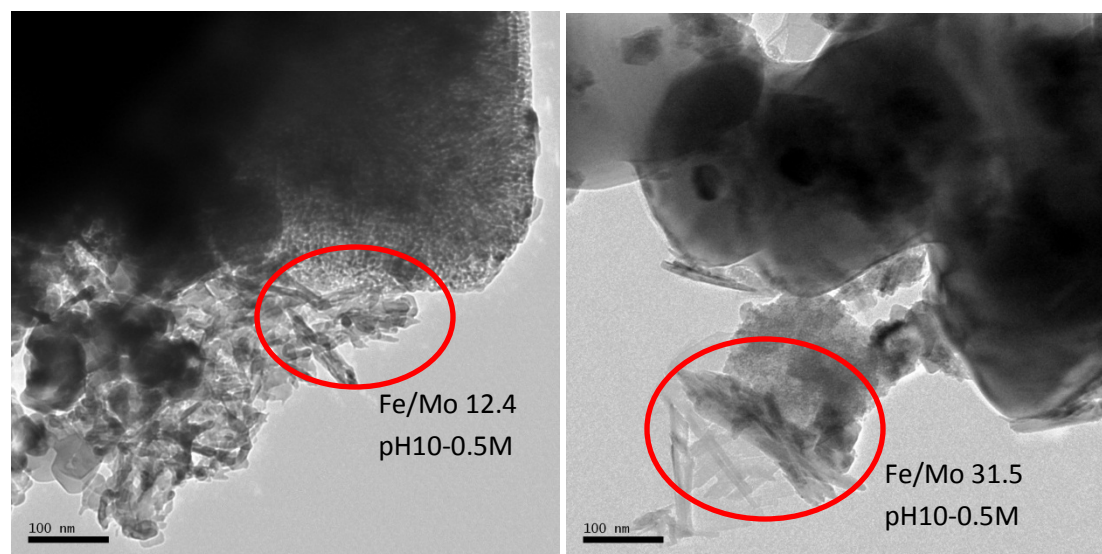


Fig S10. TEM of Fe/Mo 12.4 and Fe/Mo 31.5 after reaction for 7 days at a target pH of 10 and a concentration of 0.5mM $\text{Fe(II)}_{(\text{aq})}$. The red outlines show the sharp nano-crystalline particles that are beginning form from the bulk FH matrix.