

Supporting information for

Fate of adsorbed arsenic during early stage sulfidization of nano-ferrihydrite

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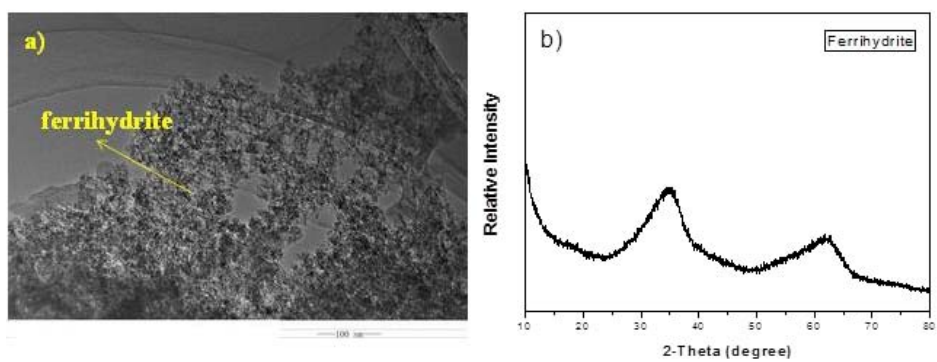


Figure. S1 TEM image (a) and XRD pattern (b) of nano-ferrihydrate

Table S1: Results of Phase identification and semi-quantitative Rietveld refinement for phase analysis of As-free and As(III)-adsorbed Fhy in presence of 0.5 mM S(-II) at different Fe/As molar ratios after 15 day reaction.

	Gt %	Lp %	Fhy%	Rexp	Rwp
As free	100	0	0	4.8	16.05
Fe/As(V) 500	100	0	0	4.63	13.76
Fe/As(V) 100	2.9	85.8	11.3	4.31	18.6
Fe/As(V) 50	0.2	64.8	35	4.4	18.56
Fe/As(III) 500	99.5	0	0.5	4.51	16.26
Fe/As(III) 100	99.6	0	0.4	4.52	11.38
Fe/As(III) 50	79.5	16.8	3.8	4.45	14.01

Phase identification and semi-quantitative Rietveld refinement

Phase identification and semi-quantitative Rietveld refinement for phase analysis was conducted with X'pert HighScore Plus software using the PAN-Inorganic and Mineral Crystal Structure Database version 2.0.23 (Table S1). In all cases, the iron oxyhydroxide used for analysis (with JCPDS #) were: 2-line Fhy (98-011-1017),¹ goethite (98-011-2696),² and lepidocrocite (98-006-5118).³ The semi-quantitative Rietveld phase conducted with the X'pert HighScore Plus software was tested for validity by analyzing a reagent grade goethite standard and refining the XRD data with unlikely multiple phases with diffraction peaks in all regions. Rietveld analysis (accuracy of $\leq 5\%$) confirmed the major phase in the test samples.

Table S2: Results of linear combination fits (LCF) for S-K-edge XANES spectra following reaction As-free and As(III)-adsorbed Fhy in presence of 0.5 mM S(-II) at different Fe/As molar ratios.

	FeS	S ⁰	As ₂ S ₃	Na ₂ SO ₃	NaS ₂ O ₃	Na ₂ SO ₄	R-factor
As free-0.25d	0	10.7	0	17.5	46.9	24.9	0.0158
As free-1d	0	0	0	5.8	43.5	50.7	0.0220
As free-15d	0	0	0	16.1	18.6	65.3	0.0061
Fe/As(V)500-0.25d	0	2.8	0	15.1	50.6	31.4	0.0137
Fe/As(V)500-1d	0	0	0	12.7	33.9	53.4	0.0142
Fe/As(V)500-15d	0	0	0	18.8	19.6	61.6	0.0142
Fe/As(V)100-0.25d	0	8.2	0	11.7	47.7	32.7	0.0141
Fe/As(V)100-1d	0	0	0	4	32	64.1	0.0113
Fe/As(V)100-15d	0	0	0	0	21.2	78.8	0.0107
Fe/As(V)50-0.25d	0	2.8	0	12.4	51.5	33.3	0.0141
Fe/As(V)50-1d	0	0	0	8.1	42.1	49.8	0.0159
Fe/As(V)50-15d	0	0	0	0	12	88	0.0053
Fe/As(III)500-0.25d	0	0	4.5	17.5	50.1	27.9	0.0184
Fe/As(III)500-1d	0	0	0	17.1	18.8	64.1	0.0140
Fe/As(III)500-15d	0	0	0	17.7	16	66.3	0.0153
Fe/As(III)100-0.25d	0	0	8.5	15.8	62.4	13.3	0.0297
Fe/As(III)100-1d	0	0	0	8.9	69.2	21.9	0.0269
Fe/As(III)100-15d	0	0	6.6	9.1	13.2	71	0.0640
Fe/As(III)50-0.25d	0	0	27.7	9.4	62.1	0.8	0.0349
Fe/As(III)50-1d	0	0	15.3	6.3	73.9	4.5	0.0747
Fe/As(III)50-15d	0	0	25.9	6.9	54.9	12.3	0.0873

Table S3: Results of linear combination fits (LCF) for As-K-edge XANES spectra following reaction As-free and As(III)-adsorbed Fhy in presence of 0.5 mM S(-II) at different Fe/As molar ratios.

	As(V)	As(III)	R-factor
Fe/As(V)50-15d	100	0	0.1094
Fe/As(III)100-1d	30.4	69.6	0.0082
Fe/As(III)100-15d	45.7	54.3	0.0410
Fe/As(III)50-1d	31.5	68.5	0.0126
Fe/As(III)50-15d	50.7	49.3	0.0479

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