

Supplementary Information

^2H and ^{27}Al Solid-State NMR Study of the Local Environments in Al-Doped 2-Line Ferrihydrite, Goethite, and Lepidocrocite

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Configuration	Unscaled ^{27}Al shifts / 10^5 ppm		Scaled ^{27}Al shifts / ppm	
	Al atom 1	Al atom 2	Al atom 1	Al atom 2
Goethite				
Single	3.02		662	
1NN	1.67	1.67	366	366
2NN	3.70	3.70	813	813
3NN	2.42	2.58	532	565
Lepidocrocite				
Single	5.20		4760	
1NN	5.20	5.21	4690	4700
2NN	5.27	5.33	4750	4810
3NN	1.61	1.60	1450	1450

Table S1. Calculated ^{27}Al unscaled and scaled hyperfine shifts for Al-doped goethite and lepidocrocite configurations with 35% Hartree-Fock exchange (results for 20% HF exchange displayed in main manuscript).

(a) **Goethite**, pristine ^2H shift: 42 ppm (b) **Lepidocrocite**, pristine ^2H shift: 380 ppm

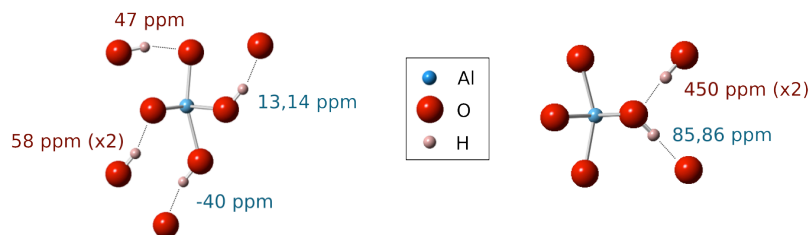


Figure S1. Results from the first principles calculation of the ^2H hyperfine shifts using 35% Hartree-Fock exchange in (a) goethite and (b) lepidocrocite. The shifts for the unique ^2H nucleus in the pristine material are quoted while the individual ^2H shifts for sites surrounding a single Al dopant are labeled on the associated structures.

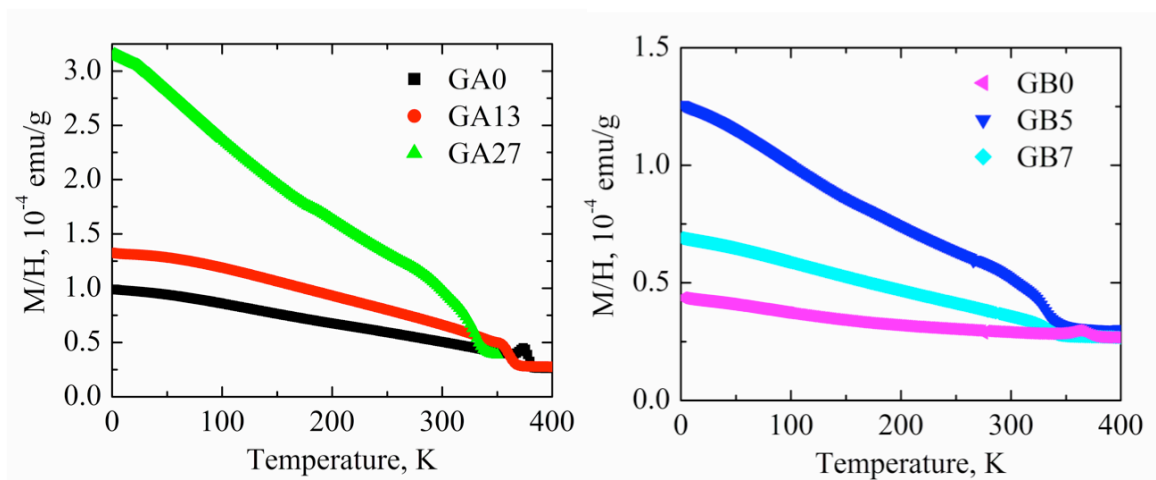


Figure S2. Magnetization curves for all of the goethite series as a function of temperature.