

---SUPPLEMENTARY INFORMATION---

**Identification of Non-Traditional Coordination Environments for Iron Ions in Nickel Hydroxide Lattices**

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**Table S1.** Charges passed in the pre-catalytic cathodic redox peaks in cyclic voltammograms that are used to normalize current flow in electrochemical experiments.

	<b>Water (mC)</b>	<b>Formamide (mC)</b>
0% Fe	22.61	23.11
0.5% Fe	22.99	22.58
2.7% Fe	23.43	16.34
5.7% Fe	18.81	13.36
8.7% Fe	15.60	26.05
11.9% Fe	12.43	19.03
15.3% Fe	2.838	4.982
18.9% Fe	12.12	18.84
21.4% Fe	18.32	12.44

**Table S2.** Location of the Ni and Fe K-edges from XANES spectra on the two sample series.<sup>a</sup>

	<b>Formamide Sample Series</b>		<b>Water Sample Series</b>	
	<b>Ni K-edge</b>	<b>Fe K-edge</b>	<b>Ni K-edge</b>	<b>Fe K-edge</b>
0% Fe	8341.33 (0.002)		8341.39 (0.035)	
5% Fe	8341.38 (0.004)	7124.97 (0.054)	8341.26 (0.006)	7124.58 (0.067)
10% Fe	8341.39 (0.068)	7124.63 (0.041)	8341.53 (0.006)	7124.56 (0.024)
15% Fe	8341.46 (0.004)	7124.47 (0.025)	8341.38 (0.006)	7124.39 (0.042)
20% Fe	8341.44 (0.027)	7124.33 (0.025)	8341.34 (0.009)	7124.30 (0.023)
25% Fe	8341.49 (0.004)	7124.17 (0.011)	8341.42 (0.003)	7124.06 (0.011)
30% Fe	8341.48 (0.009)	7124.93 (0.014)	8341.43 (0.009)	7123.90 (0.026)

<sup>a</sup> estimated errors are standard deviation from all individual spectra per sample

**Table S3.** EXAFS preliminary simulation parameters of Ni K-edge of water series

<b>Sample</b>	$\sigma_o^2$	$\sigma_{Ni}^2$	$E_o$	$R_{Ni-O}$	$R_{Ni-M}$	<b>R-factor</b>
0% Fe	0.0057 (0.0005)	0.0071 (0.0005)	8.174 (0.476)	2.065 (0.0057)	3.116 (0.0053)	0.006
5% Fe	0.0069 (0.0005)	0.0077 (0.0004)	8.328 (0.421)	2.068 (0.0052)	3.116 (0.0048)	0.007
10% Fe	(0.0053) (0.0006)	0.0070 (0.0006)	8.290 (0.599)	2.065 (0.007)	3.110 (0.007)	0.010
15% Fe	0.0061 (0.0005)	0.0073 (0.0005)	8.097 (0.516)	2.063 (0.006)	3.103 (0.005)	0.007
20% Fe	0.0073 (0.0006)	0.0078 (0.0005)	8.133 (0.582)	2.062 (0.007)	3.098 (0.006)	0.014
25% Fe	0.0065 (0.0008)	0.0075 (0.0008)	8.329 (0.767)	2.065 (0.009)	3.095 (0.008)	0.019
30% Fe	0.0058 (0.0007)	0.0074 (0.0007)	8.146 (0.679)	2.061 (0.008)	3.090 (0.007)	0.012

$N_{Ni-O}, N_{Ni-M} = 6, S_o^2 = 0.75, E_{o,Ni} = 8333 \text{ eV}$

**Table S4.** EXAFS preliminary simulation parameters of Fe K-edge of water series

<b>Sample</b>	$\sigma_o^2$	$\sigma_{Fe}^2$	$E_o$	$R_{Fe-O}$	$R_{Fe-M}$	<b>R-factor</b>
5% Fe	0.0039 (0.0006)	0.0065 (0.0007)	12.362 (0.555)	2.000 (0.007)	3.112 (0.007)	0.015
10% Fe	0.0041 (0.0005)	0.0068 (0.0007)	12.342 (0.492)	2.000 (0.006)	3.108 (0.006)	0.013
15% Fe	0.0047 (0.0005)	0.0071 (0.0007)	12.163 (0.500)	1.998 (0.006)	3.100 (0.007)	0.015
20% Fe	0.0062 (0.0006)	0.0078 (0.0007)	12.181 (0.521)	1.996 (0.007)	3.090 (0.007)	0.023
25% Fe	0.0080 (0.0008)	0.0090 (0.0009)	12.054 (0.641)	1.991 (0.008)	3.086 (0.009)	0.027
30% Fe	0.0089 (0.0008)	0.0103 (0.0012)	11.883 (0.715)	1.989 (0.010)	3.076 (0.011)	0.044

$N_{Fe-O}, N_{Fe-M} = 6, S_o^2 = 0.80, E_{o,Fe} = 7112 \text{ eV}$

**Table S5.** EXAFS preliminary simulation parameters of Ni K-edge of formamide series

<b>Sample</b>	$\sigma_{\text{O}}^2$	$\sigma_{\text{Ni}}^2$	$E_0$	$R_{\text{Ni-O}}$	$R_{\text{Ni-M}}$	<b>R-factor</b>
0% Fe	0.0061 (0.0005)	0.0074 (0.0005)	8.196 (0.497)	2.064 (0.006)	3.118 (0.005)	0.012
5% Fe	0.0060 (0.0005)	0.0074 (0.0005)	8.005 (0.501)	2.061 (0.006)	3.110 (0.005)	0.009
10% Fe	0.0057 (0.0006)	0.0072 (0.0006)	7.996 (0.598)	2.058 (0.007)	3.102 (0.006)	0.013
15% Fe	0.0056 (0.0007)	0.0071 (0.0007)	8.185 (0.648)	2.059 (0.008)	3.099 (0.007)	0.013
20% Fe	0.0058 (0.0007)	0.0070 (0.0007)	8.100 (0.686)	2.059 (0.008)	3.095 (0.007)	0.013
25% Fe	0.0052 (0.0007)	0.0070 (0.0007)	8.145 (0.717)	2.057 (0.008)	3.092 (0.008)	0.016
30% Fe	0.0050 (0.0007)	0.0074 (0.0008)	8.312 (0.736)	2.060 (0.009)	3.092 (0.009)	0.020

$N_{\text{Ni-O}}, N_{\text{Ni-M}} = 6, S_0^2 = 0.75, E_{0,\text{Ni}} = 8333 \text{ eV}$

**Table S6.** EXAFS preliminary simulation parameters of Fe K-edge of formamide series

<b>Sample</b>	$\sigma_{\text{O}}^2$	$\sigma_{\text{Fe}}^2$	$E_0$	$R_{\text{Fe-O}}$	$R_{\text{Fe-M}}$	<b>R-factor</b>
5% Fe	0.0037 (0.0006)	0.0053 (0.0006)	13.085 (0.521)	2.002 (0.006)	3.111 (0.006)	0.012
10% Fe	0.0042 (0.0005)	0.0051 (0.0005)	12.755 (0.473)	1.998 (0.006)	3.096 (0.005)	0.010
15% Fe	0.0049 (0.0006)	0.0060 (0.0006)	12.623 (0.561)	1.997 (0.007)	3.089 (0.006)	0.013
20% Fe	0.0065 (0.0007)	0.0070 (0.0007)	12.704 (0.590)	1.998 (0.008)	3.089 (0.007)	0.017
25% Fe	0.0079 (0.0007)	0.0082 (0.0007)	12.472 (0.560)	1.994 (0.007)	3.081 (0.007)	0.015
30% Fe	0.0093 (0.0009)	0.0092 (0.0010)	12.623 (0.708)	1.995 (0.010)	3.077 (0.009)	0.023

$N_{\text{Fe-O}}, N_{\text{Fe-M}} = 6, S_0^2 = 0.80, E_{0,\text{Fe}} = 7112 \text{ eV}$

**Table S7.** EXAFS final simulation parameters of Ni K-edge of water series

<b>Sample</b>	$\sigma_{\text{O}}^2$	$\sigma_{\text{Ni}}^2$	$R_{\text{Ni-O}}$	$R_{\text{Ni-M}}$	$R\text{-factor}$
0% Fe	0.0058 (0.0005)	0.0072 (0.0005)	2.065 (0.003)	3.116 (0.004)	0.0064
5% Fe	0.0069 (0.0004)	0.0077 (0.0004)	2.066 (0.002)	3.115 (0.003)	0.0072
10% Fe	0.0053 (0.0006)	0.0070 (0.0006)	2.063 (0.003)	3.109 (0.005)	0.010
15% Fe	0.0061 (0.0005)	0.0073 (0.0005)	2.063 (0.003)	3.104 (0.004)	0.008
20% Fe	0.0073 (0.0006)	0.0078 (0.0005)	2.062 (0.003)	3.098 (0.004)	0.014
25% Fe	0.0064 (0.0008)	0.0075 (0.0007)	2.063 (0.004)	3.093 (0.006)	0.018
30% Fe	0.0058 (0.0007)	0.0074 (0.0006)	2.061 (0.004)	3.090 (0.005)	0.012

$N_{\text{Ni-O}}, N_{\text{Ni-M}} = 6, S_0^2 = 0.75, E_{0,\text{Ni}} = 8333 \text{ eV}, E_0 = 8.15 \text{ eV}$

**Table S8.** EXAFS final simulation parameters of Fe K-edge of water series

<b>Sample</b>	$\sigma_{\text{O}}^2$	$\sigma_{\text{Fe}}^2$	$R_{\text{Fe-O}}$	$R_{\text{Fe-M}}$	$R\text{-factor}$
5% Fe	0.0039 (0.0006)	0.0065 (0.0007)	1.998 (0.003)	3.110 (0.006)	0.016
10% Fe	0.0041 (0.0005)	0.0068 (0.0006)	1.998 (0.003)	3.106 (0.005)	0.014
15% Fe	0.0047 (0.0005)	0.0071 (0.0006)	1.998 (0.003)	3.100 (0.005)	0.015
20% Fe	0.0062 (0.0006)	0.0078 (0.0007)	1.995 (0.003)	3.090 (0.005)	0.023
25% Fe	0.0080 (0.0007)	0.0090 (0.0009)	1.993 (0.004)	3.087 (0.007)	0.027
30% Fe	0.0089 (0.0008)	0.0103 (0.0011)	1.992 (0.004)	3.078 (0.008)	0.042

$N_{\text{Fe-O}}, N_{\text{Fe-M}} = 6, S_0^2 = 0.80, E_{0,\text{Fe}} = 7112, E_0: 12.15 \text{ eV}$

**Table S9.** EXAFS final simulation parameters of Ni K-edge of formamide series

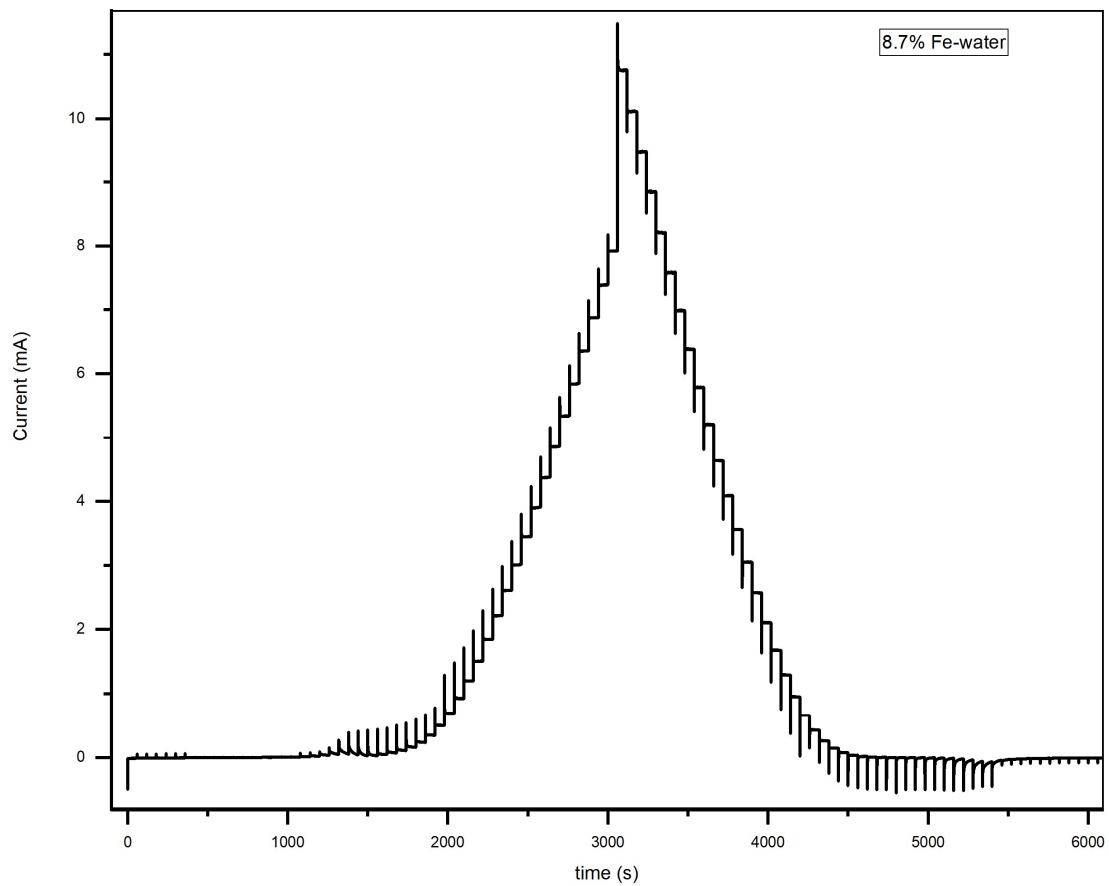
<b>Sample</b>	$\sigma_{\text{O}}^2$	$\sigma_{\text{Ni}}^2$	$R_{\text{Ni-O}}$	$R_{\text{Ni-M}}$	$R\text{-factor}$
0% Fe	0.061 (0.0005)	0.0074 (0.0005)	2.064 (0.003)	3.118 (0.004)	0.012
5% Fe	0.006 (0.0005)	0.0074 (0.0005)	2.062 (0.003)	3.111 (0.004)	0.009
10% Fe	0.0057 (0.0006)	0.0072 (0.0006)	2.060 (0.003)	3.104 (0.005)	0.013
15% Fe	0.0056 (0.0006)	0.0071 (0.0006)	2.059 (0.004)	3.099 (0.005)	0.013
20% Fe	0.0058 (0.0007)	0.0070 (0.0007)	2.059 (0.004)	3.095 (0.005)	0.014
25% Fe	0.0052 (0.0007)	0.0069 (0.0007)	2.057 (0.004)	3.092 (0.006)	0.016
30% Fe	0.0050 (0.0007)	0.0075 (0.0008)	2.058 (0.004)	3.090 (0.007)	0.020

$N_{\text{Ni-O}}, N_{\text{Ni-M}} = 6, S_0^2 = 0.75, E_{0,\text{Ni}} = 8333 \text{ eV}, E_0 = 8.15 \text{ eV}$

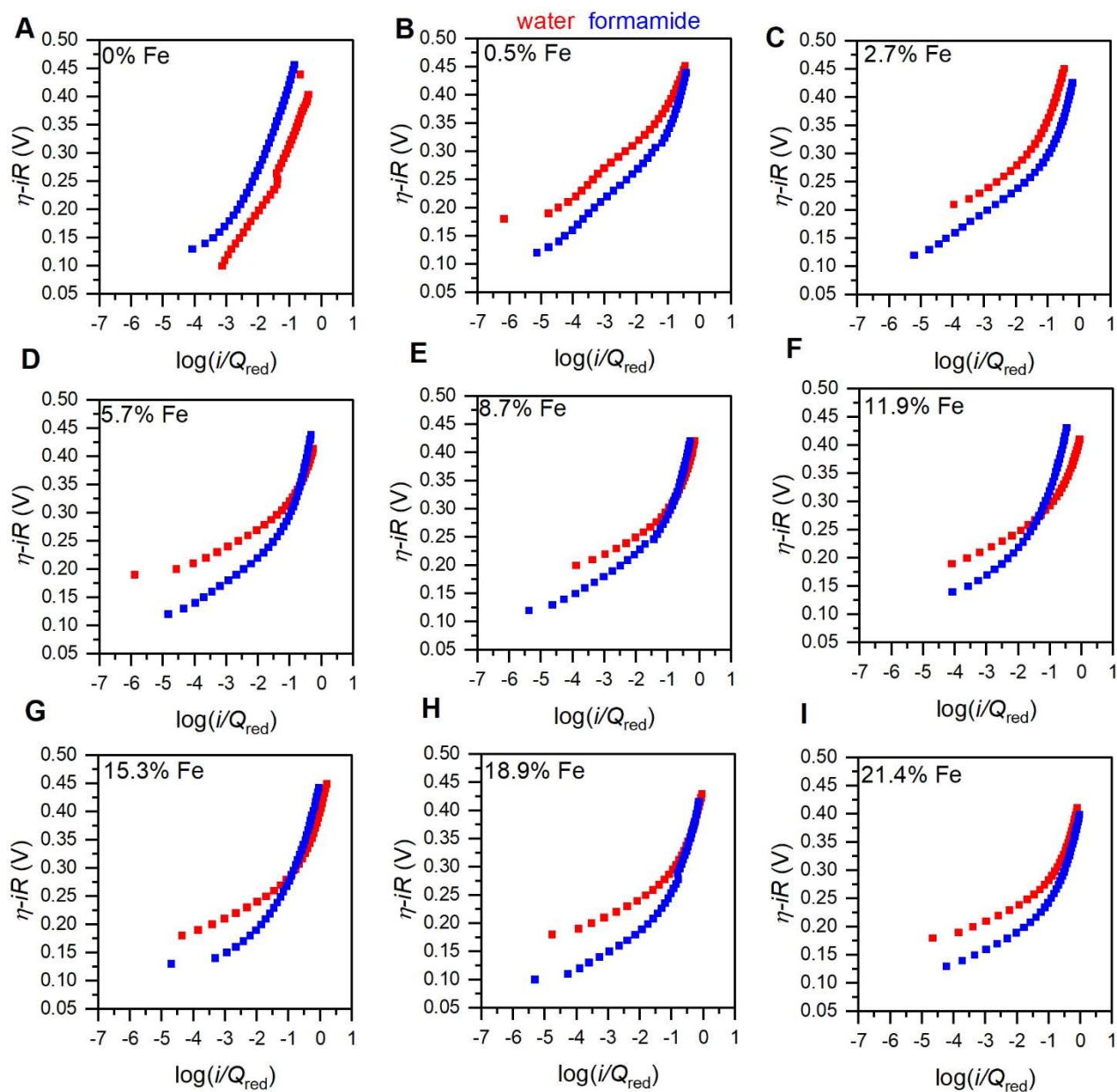
**Table S10.** EXAFS final simulation parameters of Fe K-edge of formamide series

<b>Sample</b>	$\sigma_{\text{O}}^2$	$\sigma_{\text{Fe}}^2$	$R_{\text{Fe-O}}$	$R_{\text{Fe-M}}$	$R\text{-factor}$
5% Fe	0.0036 (0.0006)	0.0053 (0.0006)	1.992 (0.004)	3.104 (0.005)	0.017
10% Fe	0.0042 (0.0005)	0.0052 (0.0005)	1.992 (0.003)	3.092 (0.004)	0.012
15% Fe	0.0049 (0.0006)	0.0060 (0.0006)	1.992 (0.003)	3.085 (0.005)	0.014
20% Fe	0.0065 (0.0007)	0.0070 (0.0007)	1.992 (0.004)	3.085 (0.006)	0.019
25% Fe	0.0078 (0.0006)	0.0082 (0.0007)	1.990 (0.003)	3.078 (0.005)	0.015
30% Fe	0.0092 (0.0008)	0.0092 (0.0009)	1.989 (0.004)	3.073 (0.007)	0.024

$N_{\text{Fe-O}}, N_{\text{Fe-M}} = 6, S_0^2 = 0.80, E_{0,\text{Fe}} = 7112, E_0: 12.15 \text{ eV}$

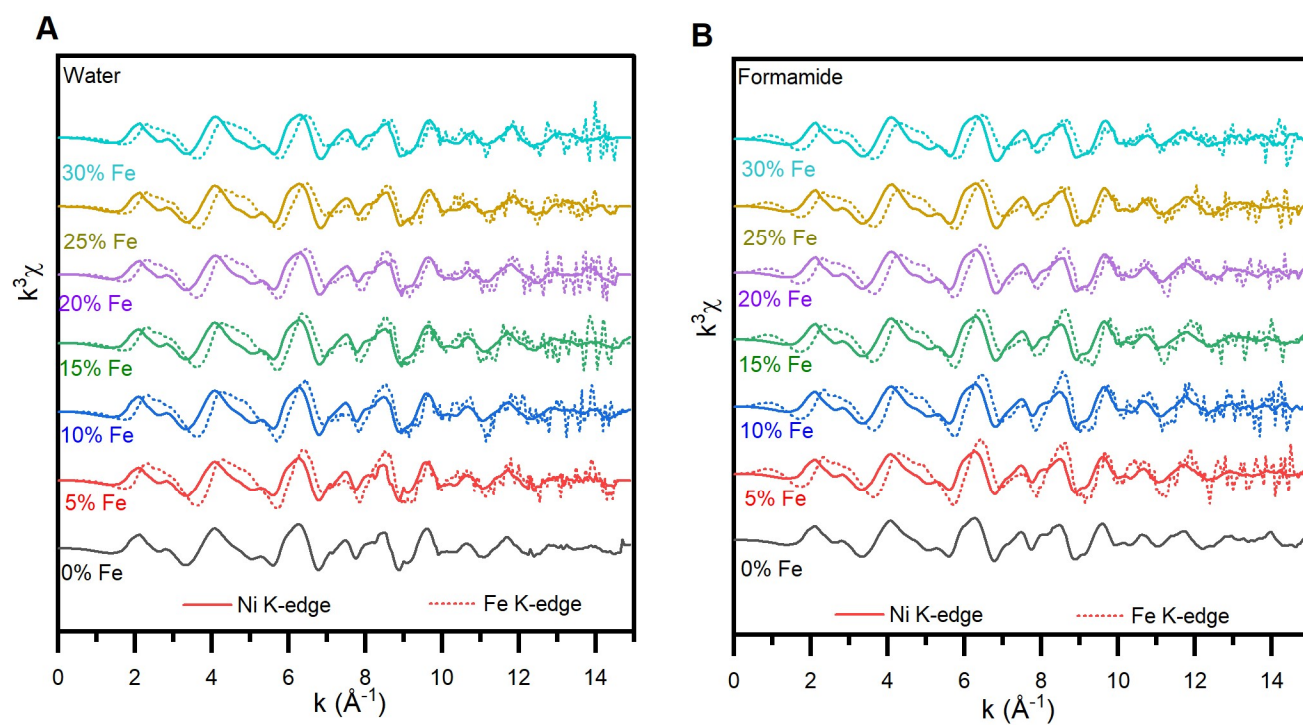


**Figure S1.** Sample chronoamperometry experiment to identify steady-state electrokinetic parameters of the materials.

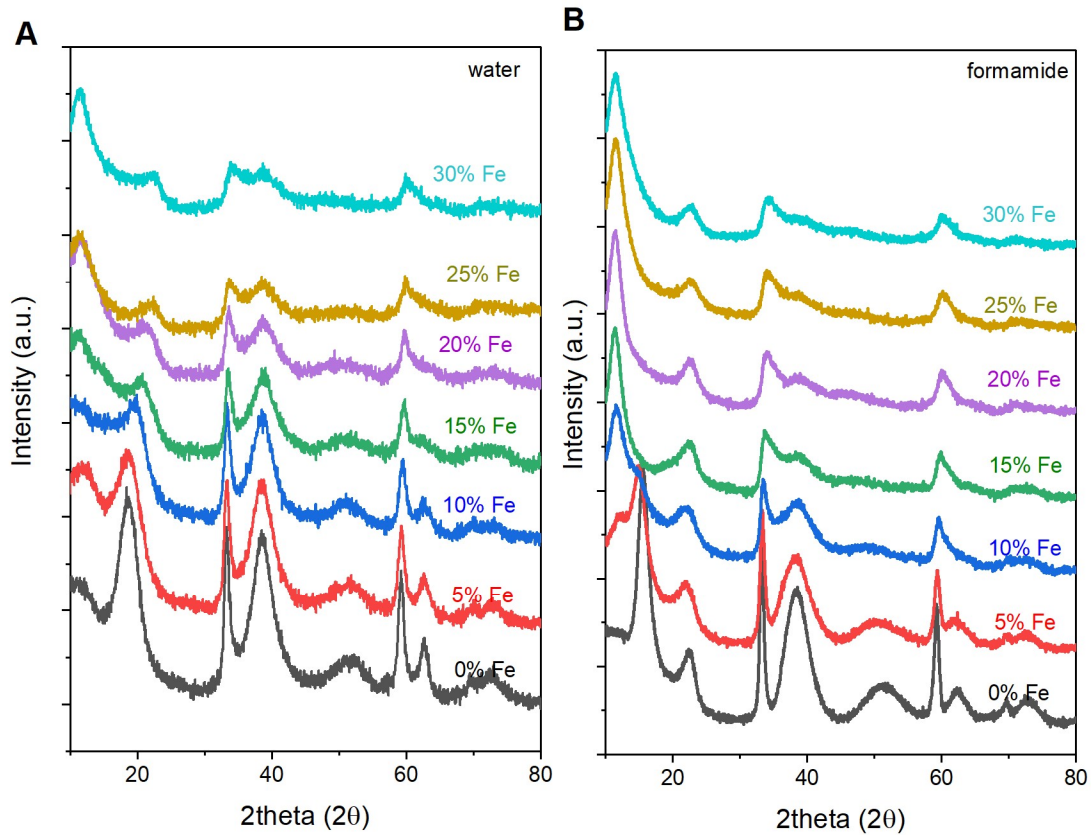


**Figure S2.** Electron transfer kinetics of water (red) and formamide(blue) series. Synthesis dependent Tafel plots for (A) 0% Fe, (B) 0.5% Fe, (C) 2.7% Fe, (D) 5.7% Fe, (E) 8.7% Fe, (F)11.9% Fe, (G) 15.3% Fe, (H) 18.9% Fe, (I) 21.4% Fe.

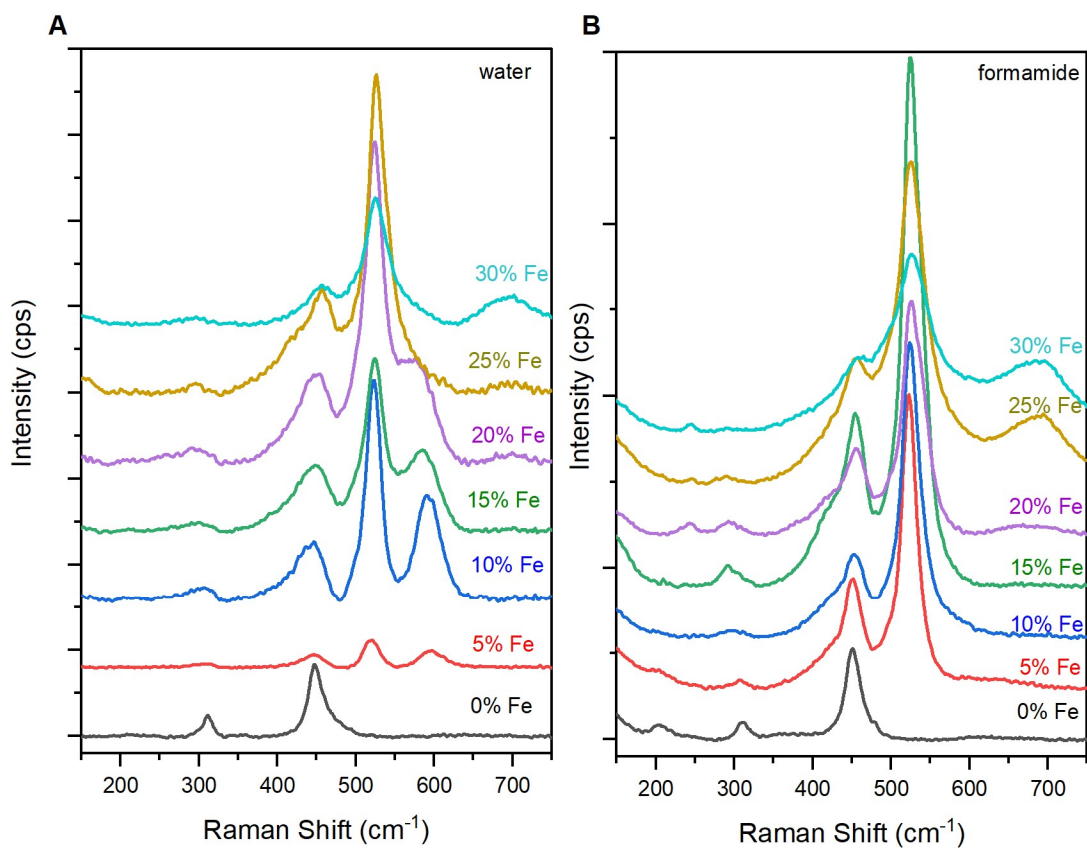




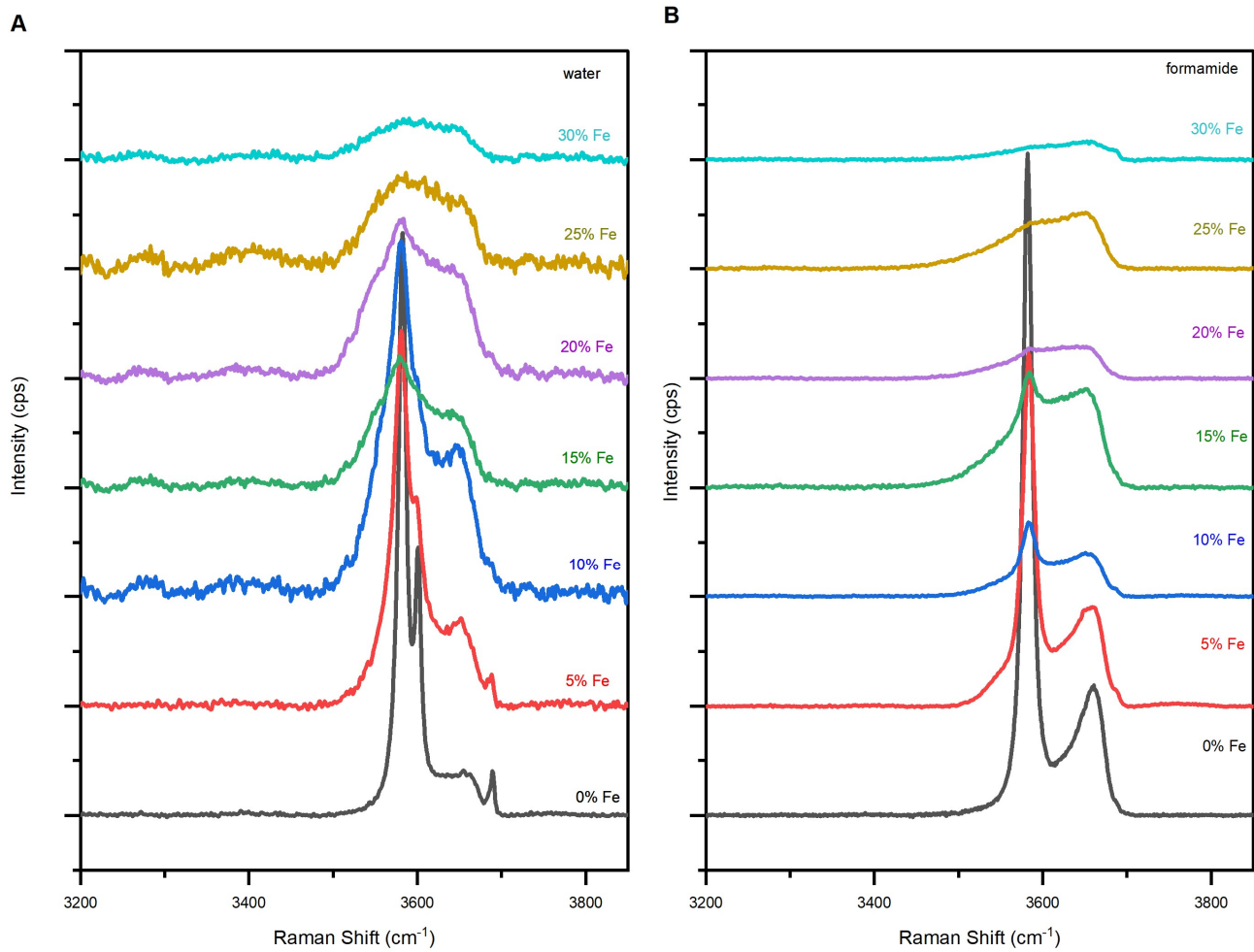
**Figure S3:** k-space X-ray absorption fine-structure spectra of water (A) and formamide (B) series at the Ni K-edge (solid lines) and Fe K-edge (dotted lines).



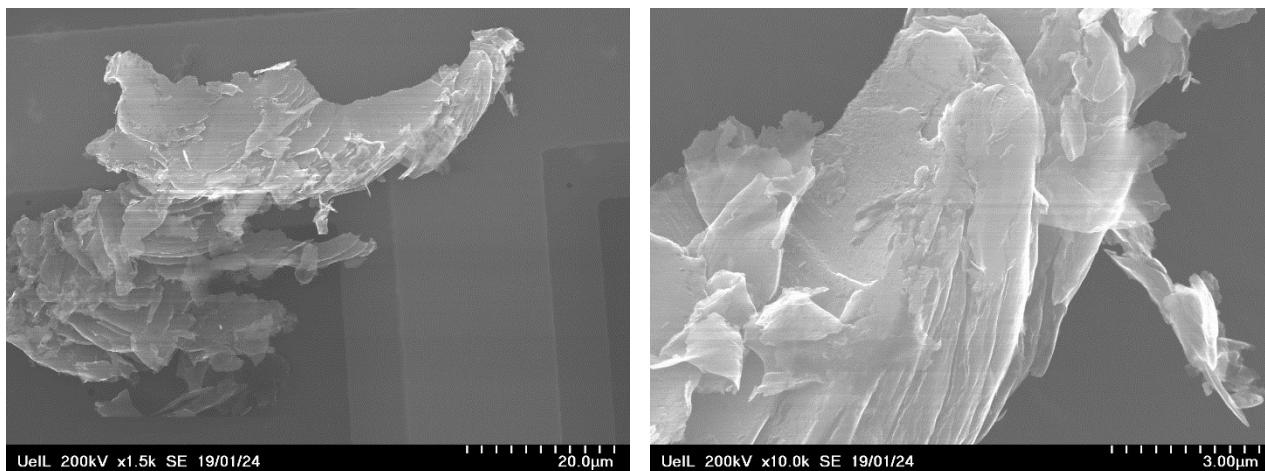
**Figure S4:** XRD patterns of the samples used in EXAFS analysis. (A) water series (B) formamide series



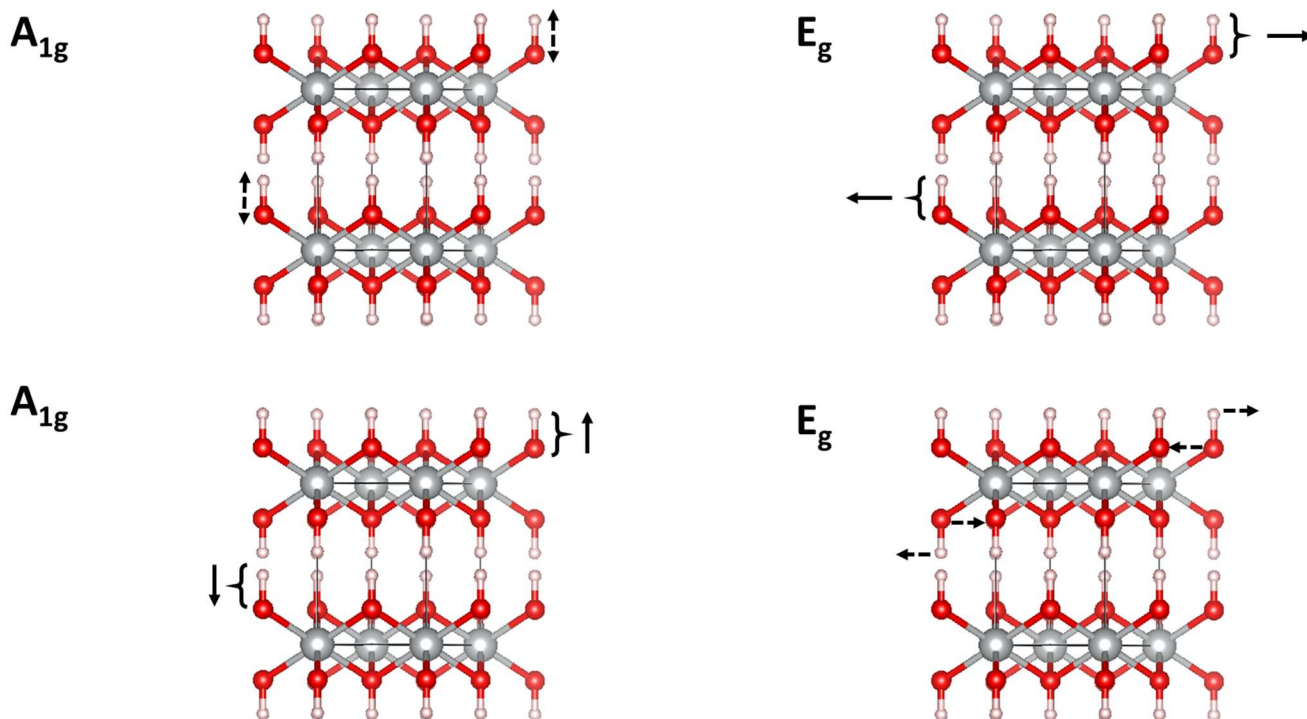
**Figure S5:** Low wavenumber Raman spectra on  $\text{Fe}_x\text{Ni}_{1-x}(\text{OH})_2$  samples used in EXAFS analysis. Data for the (A) water and (B) formamide sample series.



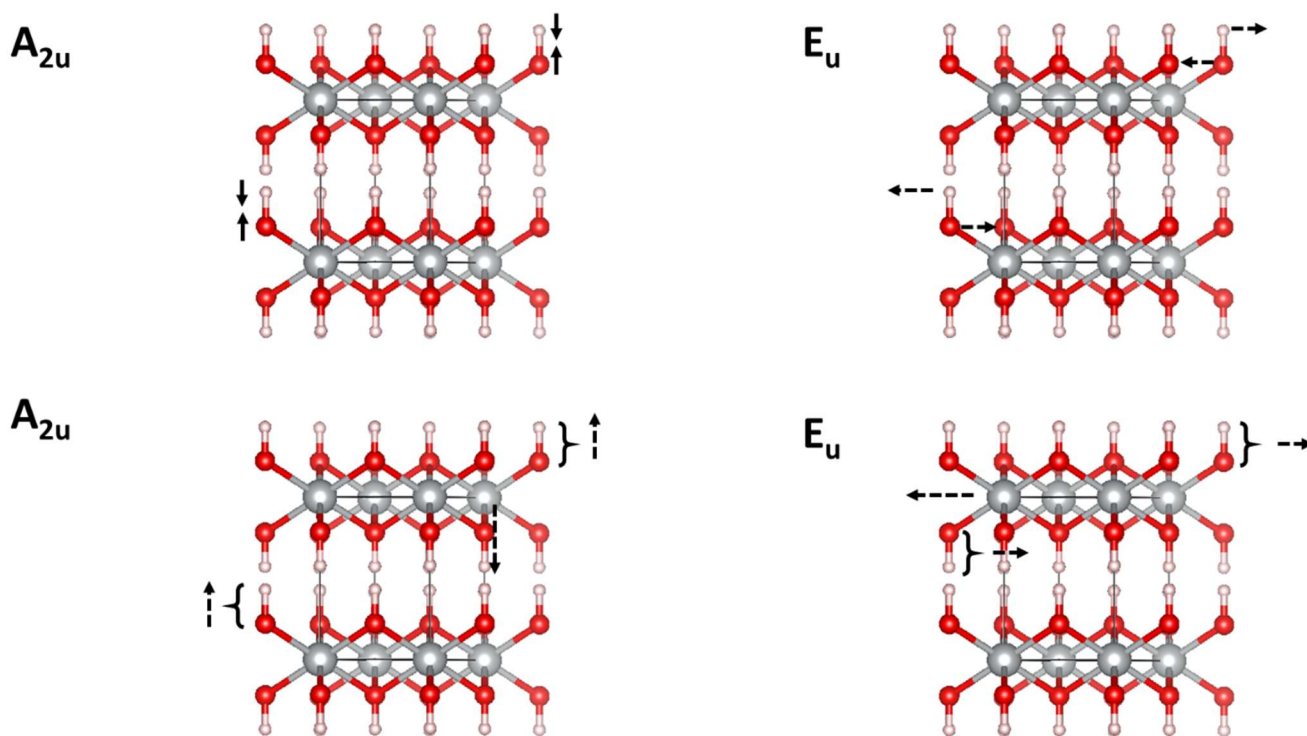
**Figure S6:** High wavenumber Raman spectra on  $\text{Fe}_x\text{Ni}_{1-x}(\text{OH})_2$  samples used in EXAFS analysis. Data for the (A) water and (B) formamide sample series.



**Figure S7:** Electron micrographs obtained on  $\text{Fe}_{0.214}\text{Ni}_{0.786}(\text{OH})_2$  synthesized in the presence of formamide.



**Figure S8:** Raman active modes of brucite ( $\beta\text{-Ni(OH)}_2$ ). Figure is adapted from<sup>1</sup>



**Figure S9:** Infrared active modes of brucite ( $\beta\text{-Ni(OH)}_2$ ). Figure is adapted from<sup>1</sup>

**REFERENCES:**

- (1) Bantignies, J. L.; Deabate, S.; Righi, A.; Rols, S.; Hermet, P.; Sauvajol, J. L.; Henn, F. New Insight into the Vibrational Behavior of Nickel Hydroxide and Oxyhydroxide Using Inelastic Neutron Scattering, Far/Mid-Infrared and Raman Spectroscopies. *Journal of Physical Chemistry C* **2008**, *112* (6), 2193–2201. <https://doi.org/10.1021/jp075819e>.