

Supporting Information for

“Superior Oxygen Evolution Reaction Activities of Highly Crystalline Ni_{1-x}Fe_x-LDH (0.20 ≤ x ≤ 0.51) Synthesized Using Soft Chemistry”

Atsunori Ikezawa*, Shinji Koito, and Hajime Arai

Department of Chemical Science and Engineering

Tokyo Institute of Technology

4259 Nagatsuta-cho, Midori-ku, Yokohama, Kanagawa, 226-8501, Japan

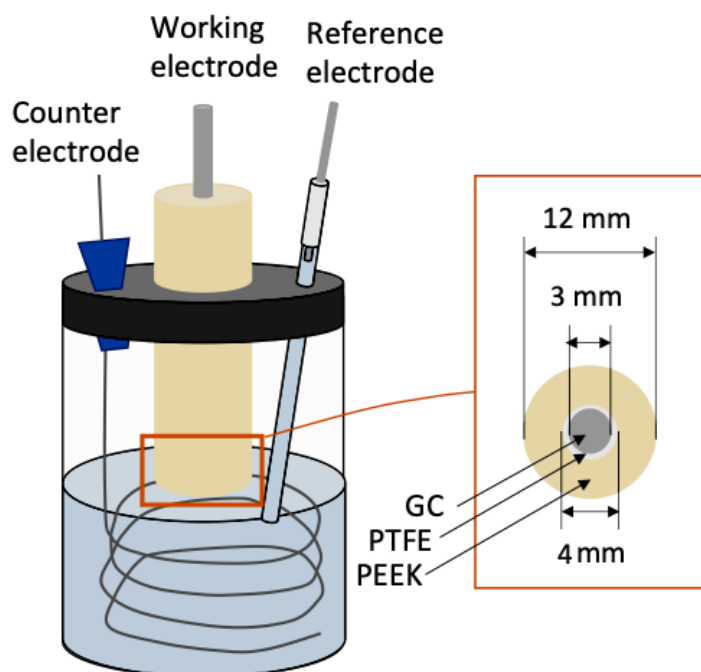
E-mail: ikezawa.a.aa@m.titech.ac.jp

Table S1. Molar ratios of the reactants for the solid phase synthesis of $\text{NaNi}_{1-x}\text{Fe}_x\text{O}_2$

Sample	Molar ratio		
	Na_2O_2	NiO	Fe_2O_3
Soft chemistry 11 % Fe	1.1	0.9	0.1
Soft chemistry 20 % Fe	1.1	0.8	0.2
Soft chemistry 32 % Fe	1.1	0.7	0.3
Soft chemistry 41 % Fe	1.1	0.6	0.4
Soft chemistry 51 % Fe	1.1	0.5	0.5

Table S2. Concentrations of the reactant solutions for the coprecipitation of the NiFe-LDHs

Sample	Concentration / mol dm^{-3}			
	$\text{Ni}(\text{NO}_3)_2$	$\text{Fe}(\text{NO}_3)_3$	NaOH	Na_2CO_3
Coprecipitation 7 % Fe	0.49	0.026	0.82	0.051
Coprecipitation 9.5 % Fe	0.46	0.051	0.82	0.10
Coprecipitation 19 % Fe	0.41	0.10	0.82	0.20

**Figure S1.** A schematic of the electrochemical three-electrode cell. GC, PTFE, and, PEEK denote glassy carbon, poly(1,1,2,2-tetrafluoroethylene), polyether ether ketone, respectively.

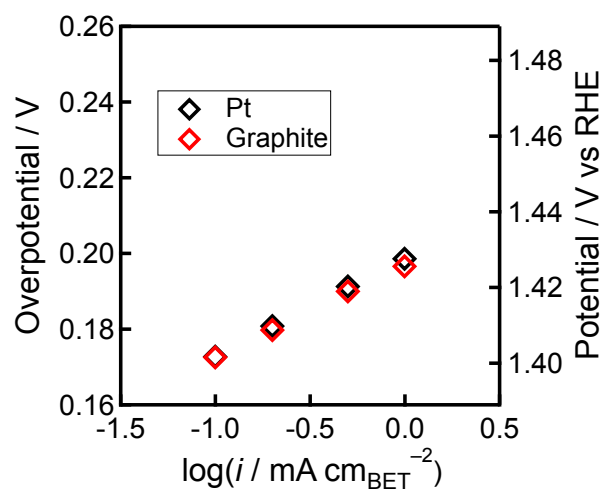


Figure S2. Comparison of Tafel plots of the soft chemistry 41 % Fe between the cell with Pt and graphite counter electrodes.

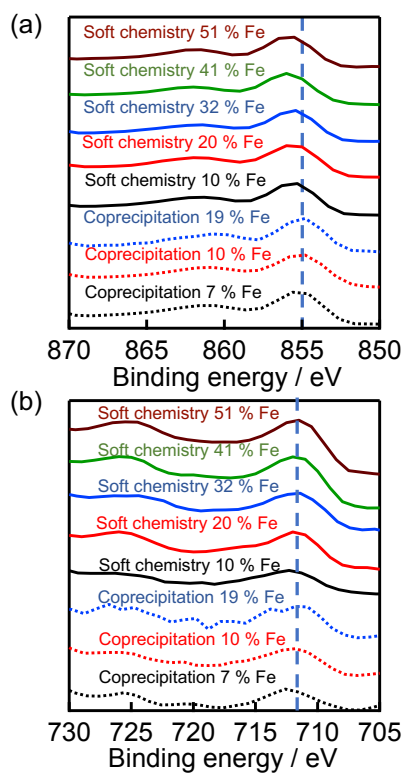


Figure S3. XPS (a) Ni2p and (b) Fe 2p spectra of the soft chemistry and coprecipitation samples.

Table S3. Current density per geometric area set at the constant current measurement

Sample	Current density / mA cm _{geo} ⁻²			
	1.0 / mA cm _{BET} ⁻²	0.5 / mA cm _{BET} ⁻²	0.2 / mA cm _{BET} ⁻²	0.1 / mA cm _{BET} ⁻²
Soft chemistry 11 % Fe	0.167	0.083	0.033	0.017
Soft chemistry 20 % Fe	0.192	0.096	0.038	0.019
Soft chemistry 32 % Fe	0.220	0.110	0.044	0.022
Soft chemistry 41 % Fe	0.219	0.109	0.044	0.022
Soft chemistry 51 % Fe	0.169	0.084	0.034	0.017
Coprecipitation 7 % Fe	1.673	0.836	0.335	0.167
Coprecipitation 9.5 % Fe	1.143	0.571	0.229	0.114
Coprecipitation 19 % Fe	0.683	0.341	0.137	0.068

Table S4. Atomic ratio of K, Na, Ni, Fe in the synthesized samples analyzed by ICP-AES

Sample	Atomic ratio / %			
	K	Na	Ni	Fe
Soft chemistry 11 % Fe	16.1	7.4	68.0	8.6
Soft chemistry 20 % Fe	10.0	8.6	61.3	15.3
Soft chemistry 32 % Fe	11.3	5.5	51.9	24.7
Soft chemistry 41 % Fe	11.2	6.9	45.3	31.2
Soft chemistry 51 % Fe	12.9	8.0	37.4	39.1
Coprecipitation 7 % Fe	0.4	5.2	71.2	5.4
Coprecipitation 9.5 % Fe	0.0	8.3	69.3	7.3
Coprecipitation 19 % Fe	0.4	4.9	61.8	14.8

Table S5. Lattice parameters obtained by the Rietveld refinement of the soft chemistry (a) 11, (b) 20, (c) 32, (d) 41, and (e) 51 %.

(a) Phase	$a / \text{\AA}$	$b / \text{\AA}$	$c / \text{\AA}$	$\alpha / ^\circ$	$\beta / ^\circ$	$\gamma / ^\circ$
$\gamma\text{-NiOOH}$	2.8337	2.8337	20.850	90.000	90.000	120.000
$\beta\text{-Ni(OH)}_2$	3.1369	3.1369	4.644	90.000	90.000	120.000
(b) Phase	$a / \text{\AA}$	$b / \text{\AA}$	$c / \text{\AA}$	$\alpha / ^\circ$	$\beta / ^\circ$	$\gamma / ^\circ$
$\gamma\text{-NiOOH}$	2.8414	2.8414	20.877	90.000	90.000	120.000
$\alpha\text{-Ni(OH)}_2$	3.0784	3.0784	23.083	90.000	90.000	120.000
(c) Phase	$a / \text{\AA}$	$b / \text{\AA}$	$c / \text{\AA}$	$\alpha / ^\circ$	$\beta / ^\circ$	$\gamma / ^\circ$
$\gamma\text{-NiOOH}$	2.8480	2.8480	21.116	90.000	90.000	120.000
$\alpha\text{-Ni(OH)}_2$	3.0734	3.0734	22.919	90.000	90.000	120.000
(d) Phase	$a / \text{\AA}$	$b / \text{\AA}$	$c / \text{\AA}$	$\alpha / ^\circ$	$\beta / ^\circ$	$\gamma / ^\circ$
$\gamma\text{-NiOOH}$	2.8563	2.8563	21.130	90.000	90.000	120.000
$\alpha\text{-Ni(OH)}_2$	3.0724	3.0724	22.744	90.000	90.000	120.000
(e) Phase	$a / \text{\AA}$	$b / \text{\AA}$	$c / \text{\AA}$	$\alpha / ^\circ$	$\beta / ^\circ$	$\gamma / ^\circ$
$\gamma\text{-NiOOH}$	2.8754	2.8754	21.062	90.000	90.000	120.000
$\alpha\text{-Ni(OH)}_2$	3.0697	3.0697	22.660	90.000	90.000	120.000

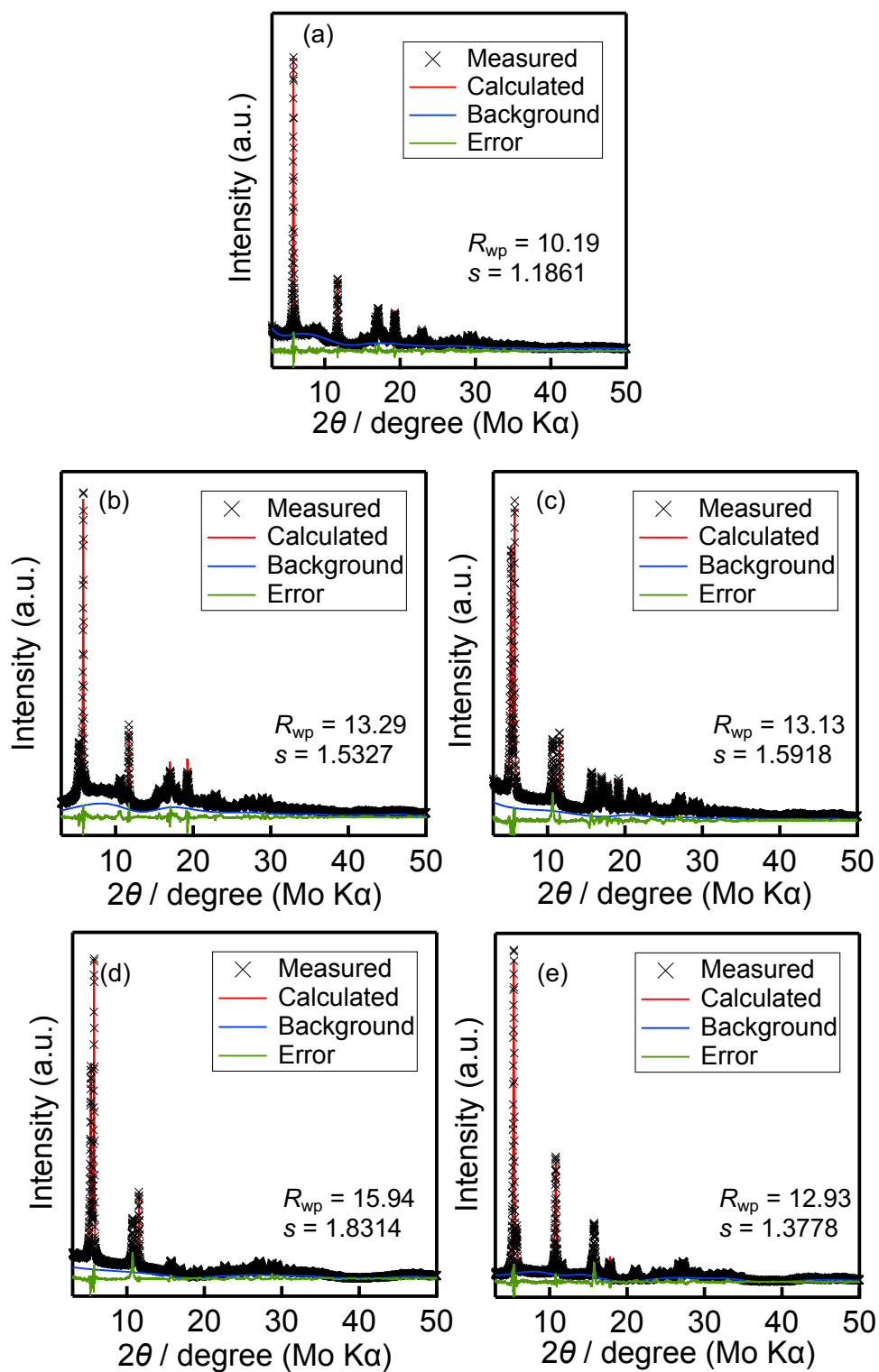


Figure S4. Results of the Rietveld refinement of the soft chemistry (a) 11, (b) 20, (c) 32, (d) 41, and (e) 51 %.

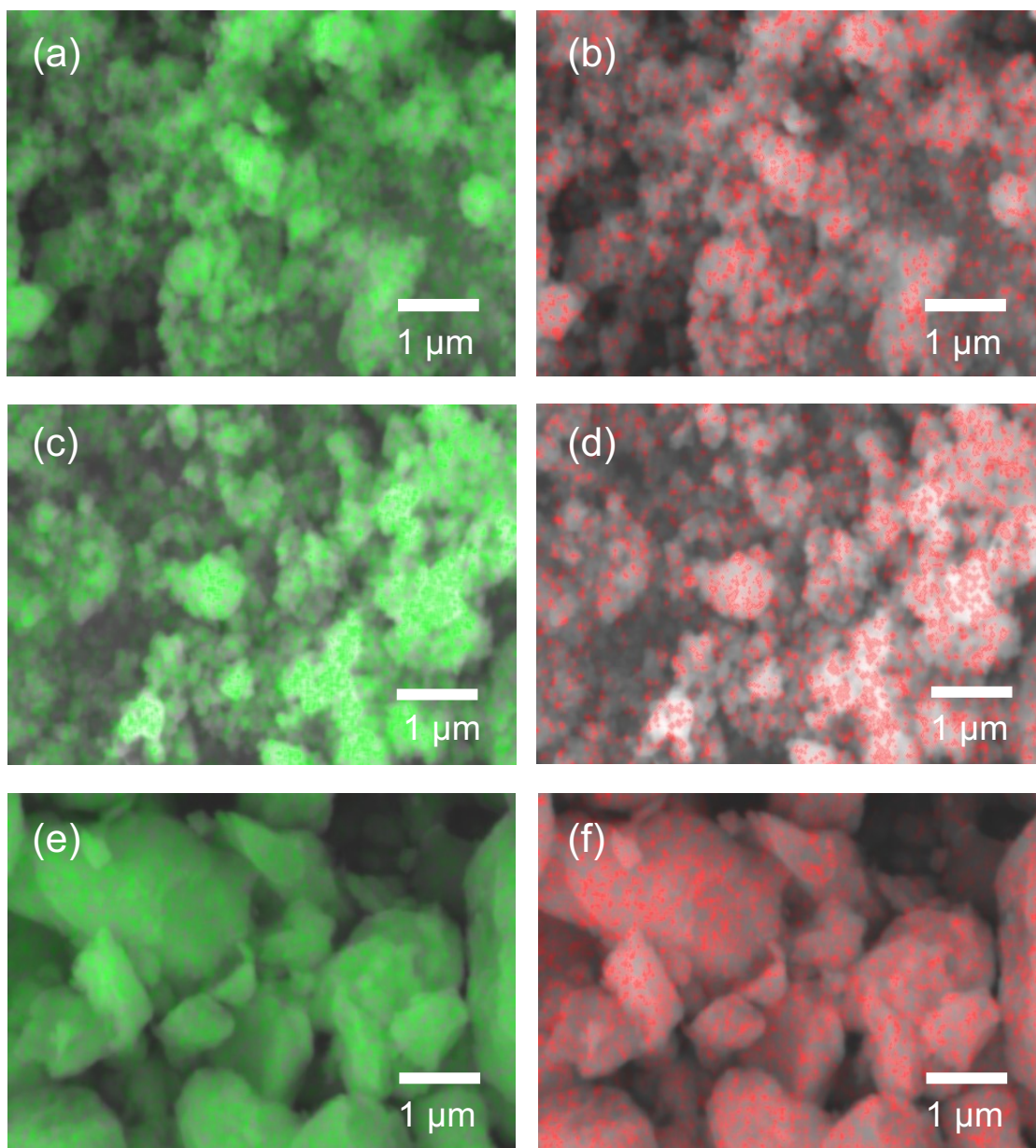


Figure S5. FE-SEM/EDS images of the coprecipitation (a,b) 7, (c,d) 9.5, and (e,f) 19 %. Blue and red regions indicate the distributions of Ni and Fe, respectively.

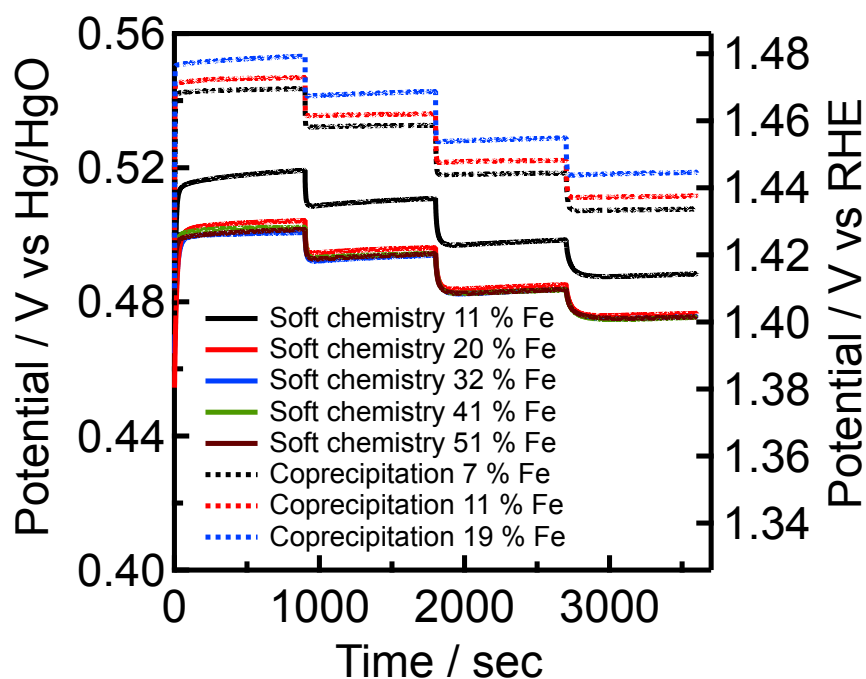


Figure S6. Chronopotentiograms of the NiFe-LDH samples during the constant current measurements at 1 mA $\text{cm}_{\text{BET}}^{-2}$, 0.50 mA $\text{cm}_{\text{BET}}^{-2}$, 0.20 mA $\text{cm}_{\text{BET}}^{-2}$, and 0.10 mA $\text{cm}_{\text{BET}}^{-2}$.

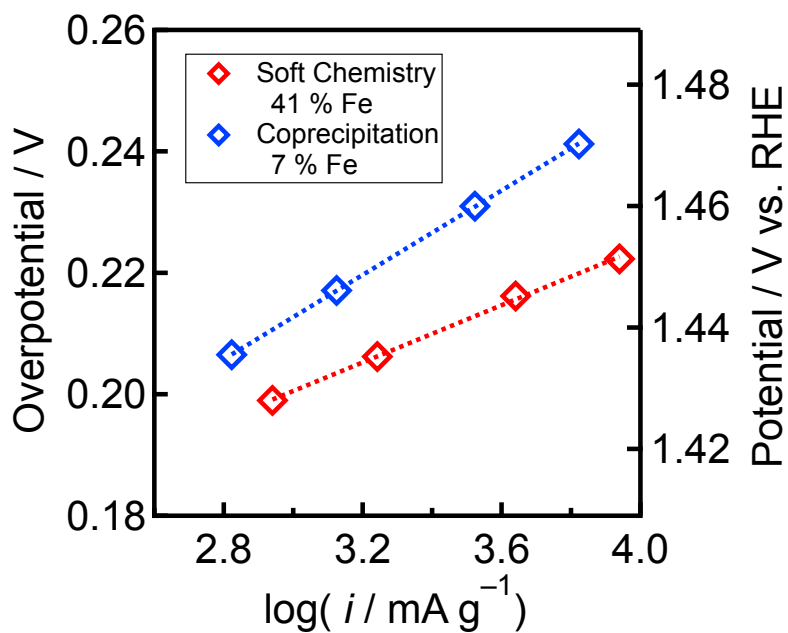


Figure S7. Comparison of Tafel plots between the soft chemistry 41 % Fe and the coprecipitation 7 % Fe at comparable current density per mass.

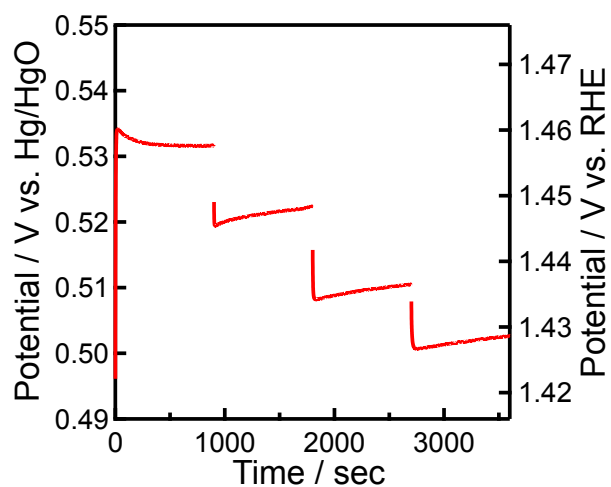


Figure S8. Chronopotentiograms of the soft chemistry 41 % Fe samples during the constant current measurements at 10 mA $\text{cm}_{\text{BET}}^{-2}$, 5.0 mA $\text{cm}_{\text{BET}}^{-2}$, 2.0 mA $\text{cm}_{\text{BET}}^{-2}$, and 1.0 mA $\text{cm}_{\text{BET}}^{-2}$.

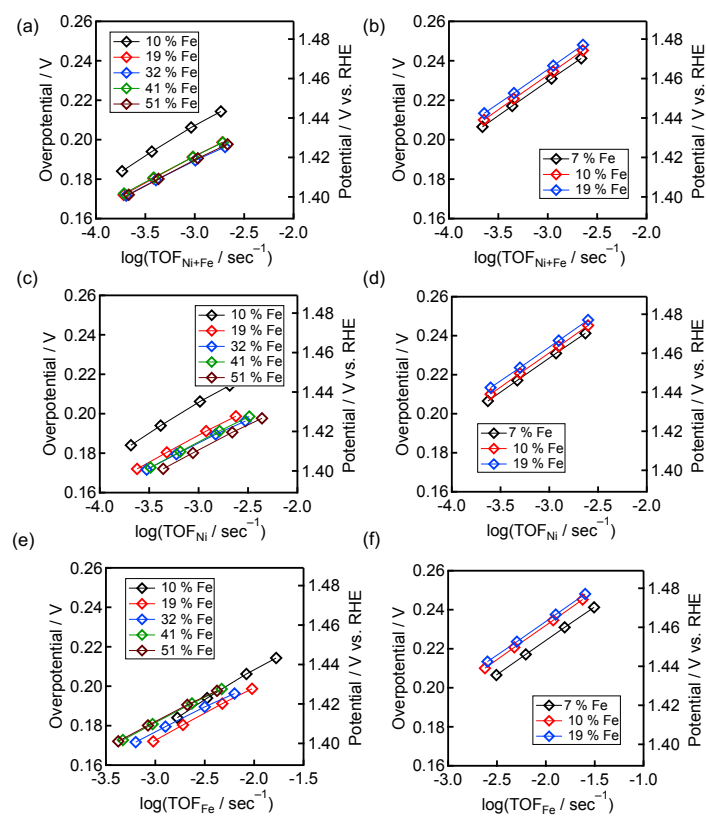


Figure S9. Relationship between overpotential and TOF of (a,b) Ni + Fe, (c,d) Ni, and (e,f) Fe in the (a,c,e) soft chemistry and (b,d,f) coprecipitation samples.