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## **Supporting Information**

## Bottom-up Synthesis of 2D Layered High-Entropy Transition Metal Hydroxides Fei Li<sup>1\*</sup>, Shi-Kuan Sun<sup>2</sup>, Yinjuan Chen<sup>3</sup>, Takashi Naka<sup>4</sup>, Takeshi Hashishin<sup>5</sup>, Jun Maruyama<sup>6</sup>, Hiroya Abe<sup>1\*</sup> <sup>1</sup> Joining and Welding Research Institute, Osaka University, Osaka 5670047, Japan <sup>2</sup> School of Material Science and Energy Engineering, Foshan University, Foshan, 528000, China <sup>3</sup> Instrumentation and Service Center for Molecular Sciences, Westlake University, Hangzhou 310024, China <sup>4</sup> National Institute for Materials Science, Ibaraki 3050047, Japan <sup>5</sup> Faculty of Advanced Science and Technology, Kumamoto University, Kumamoto, 8608555, Japan <sup>6</sup> Osaka Research Institute of Industrial Science and Technology, Osaka 5368553, Japan Corresponding authors: feili@jwri.osaka-u.ac.jp (H.A.)

Metal species	Ksp
Cr(OH) <sub>3</sub>	6.3e-31
Mn(OH) <sub>2</sub>	3e-13
Fe(OH) <sub>3</sub>	1.1e-36
Co(OH) <sub>2</sub>	5.9e-15
Ni(OH) <sub>2</sub>	1.5e-16
Zn(OH) <sub>2</sub>	3e-17

Table S1 Solubility product constants (Ksp at 25 °C).

Table S2. pH of the corresponding supernatant after hydrothermal treatment under EG-

K/N.	Interlayer	distance	for	HEH-	-2#~7#	EG-	-K/N	as s	synthesized	powders.
	-								2	

Samples	pН	Interlayer		
		distance (nm)		
HEH-2# EG-K/N	11.8±0.3	0.985		
HEH-3# EG-K/N	11.8±0.3	0.987		
HEH-4# EG-K/N	11.8±0.3	0.982		
HEH-5# EG-K/N	11.9±0.3	0.863		
HEH-6# EG-K/N	11.7±0.3	0.985		
HEH-7# EG-K/N	11.8±0.3	0.976		



Figure S1 XRD patterns of the HEH-2#~7# EG-K as synthesized powders after

hydrothermal treatment at 200  $^{\circ}\mathrm{C}$  for 2 h.



Figure S2. XRD patterns of HEH-1# as synthesized powders using EG-NaOH 200 °C hydrothermal treatment and EG-KOAC 230 °C hydrothermal treatment.



Figure S3 Elemental maps of the HEH-1# EG-K as synthesized powders on various scales. (b) and (d) show the elemental maps for selected areas in (a) as marked by solid square and circle, respectively. (c) and (e) show the elemental maps for selected areas in (b) and (d) as marked by dashed square and circle, respectively. Scale bars in (b) and (d) are 0.5 μm. Scale bar in (c) and (e) are 100 nm. Note: mix figure shows the overlapping Cr-, Fe- and Ni-signals.



Figure S4 (a)~(c) TEM images with different magnification and (d) Elemental maps of the HEH-1# EG-K/N as synthesized powders. Note: mix figure shows the overlapping Cr-, Fe- and Ni-signals. (e) AFM image of the sample.



Figure S5 TEM images with different magnification and Elemental maps of the HEH-1# $H_2O$ -K/N as synthesized powders. Note: mix figure shows the overlapping Cr-, Fe- and

Ni-signals.



Figure S6 TEM images and elemental maps for the HEH-2#~7# EG-K as synthesized

powders. Scale bars in the elemental maps are 0.5  $\mu m.$ 



Figure S7 EDS mapping for HEH-5# EG-K/N as synthesized powders showing typical

2D layered structures with uniform elemental distribution.



Figure S8 XRD patterns of the HEH-2#~7# EG-K/N powders after annealed at (a) 200

and (b) 600 °C.



Figure S9 XRD refinement of the HEH-1# (a) EG-K/N, (b)EG-K, and (c) H<sub>2</sub>O-K/N

samples after 600 °C annealing.



Figure S10 XRD patterns of the HEH-1# (a) EG-K and (b) H<sub>2</sub>O-K/N after annealing at

different temperatures.



Figure S11 TEM images and elemental maps of the HEH-1# (a~c) EG-K and (d~f)
H<sub>2</sub>O-K/N powders after annealed at 600°C. Scale bars in (c) and (d) are 2 μm. Note: mix figure shows the overlapping Cr-, Fe- and Ni-signals.



Figure S12 M-H curves for HEH-1# EG-K/N 1000°C-annealed powders and control

powders prepared by solid state reactions.



Figure S13. LSV curves of the HEH-1# as synthesized sample during 100 cyclic voltammetry

cycles.