## Engineering *rhynchostylis retusa*-like heterostructured $\alpha$ -nickel molybdate with enhanced redox properties for high-performance rechargeable asymmetric supercapacitors

Ganji Seeta Rama Raju<sup>a</sup>, Eluri Pavitra<sup>b</sup>, Goli Nagaraju<sup>c</sup>, Nilesh R. Chodankar<sup>a</sup>, Sujaya Kumar Vishwanath<sup>d</sup>, Jin Young Park<sup>e</sup>, Yun Suk Huh<sup>\*b</sup>, and Young-Kyu Han<sup>\*a</sup>

<sup>a</sup>Department of Energy and Materials Engineering, Dongguk University–Seoul, Seoul 04620, Republic of Korea

<sup>b</sup>Department of Biological Engineering, Biohybrid Systems Research Center (BSRC), Inha University, Incheon, 22212, Republic of Korea

<sup>c</sup>School of Chemistry, Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN), Trinity College Dublin, Dublin 2, Ireland

<sup>d</sup>School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798

<sup>e</sup>Department of Electrical, Electronics and Software Engineering, Pukyong National University, Busan 48573, Republic of Korea

\*Email: <u>yunsuk.huh@inha.ac.kr</u> (Y. S. Huh)

\*Email: <u>ykenergy@dongguk.edu</u> (Y.-K. Han)



**Fig. S1**. (a) XRD patterns of the as prepared NH<sub>4</sub>Ni<sub>2</sub>Mo<sub>2</sub>O<sub>8</sub>(OH)H<sub>2</sub>O as a function of TPAOH and (b) XRD patterns of  $\alpha$ -NiMoO<sub>4</sub> as a function of the amount of TPAOH,  $\alpha$ -NiMoO<sub>4</sub> phase was obtained after calcination of NH<sub>4</sub>Ni<sub>2</sub>Mo<sub>2</sub>O<sub>8</sub> at 450 C for 3 h in N2 atmosphere. (NWs, NWs+NPs, NWS+NSs+NWs+NSs+NPs were obtained when adding the amount of TPAOH 0, 10, 30, 50, and 70 µl)



**Fig. S2**. HR-SEM images of  $\alpha$ -NiMoO<sub>4</sub> nanostructures at different amounts of TPAOH (a) 10  $\mu$ l (b) 30  $\mu$ l (c) 50  $\mu$ l (d) 70  $\mu$ l, which clearly shows the influence of TPAOH on the morpholgies of  $\alpha$ -NiMoO<sub>4</sub>.

Atom	Wyckoff position	Site	Atomic coordinates			$\mathbf{I}(\mathbf{\hat{\lambda}}^2)$
Atom			x/a	y/b	z/c	U (A )
Mo1	4i	m	0.2168	0.0000	0.3659	0.0073
Mo2	4g	2	0.0000	0.2156	0.0000	0.0128
Ni1	4i	m	0.8264	0.0000	0.1613	0.0158
Ni2	4h	2	0.0000	0.1927	0.5000	0.0197
01	8j	1	0.0020	0.1555	0.2505	0.0202
02	8j	1	0.1542	0.3395	0.0714	0.0150
03	8j	1	0.3332	0.1668	0.4087	0.0458
04	4i	m	0.2186	0.0000	0.5662	0.0428
05	4i	m	0.1536	0.0000	0.0751	0.0414

**Table S1:** Crystallographic data with atomic parameters of  $\alpha$ -NiMoO<sub>4</sub>

Phase: α-NiMoO<sub>4</sub> (monoclinic) Space group: C2/m (12) Lattice constants a =9.6112 Å b= 8.7828 Å, c = 7.6728 Å, Volume: V = 590.721 Å<sup>3</sup>, α=90° β=114.21° γ= 90° R-factors:  $R_p = 2.28\%$ ,  $R_{wp} = 2.99\%$ , and  $\chi^2 = 2.098$ 



Fig. S3. Pore size distribution curves of the a-NiMoO<sub>4</sub> (a) NWs, (b) NWs+NSs, and (c) NWs+NSs+NPs)



Fig. S4. EIS spectra of  $\alpha$ -NiMoO<sub>4</sub> NWs, NWs+NSs, NWS+NSs+NPs samples measured in three-electrode system.



**Fig. S5**. (a) and (b) low and high magnification SEM images of APC, respectively, (c) CV curves of APC at different scan rates and (d) GCD curves of the APC at different current densities.