Uniform Bi-Bi$_2$O$_3$ nanoparticles/reduced graphene oxide composites for high-performance aqueous alkaline battery

Mangmang Shi[a], Mingshu Zhao*[a], Qingyang Zheng[b], Lidong Jiao[a], Zhou Su[a], Min Li[a], Xiaobo zhao[c],

Xiaoping Song[a], and Sen Yang

a. School of Physics, MOE key Laboratory for Non-Equilibrium Synthesis and Modulation of Condensed

Matter, Key Laboratory of Shaanxi for Advanced Functional Materials and Mesoscopic Physics, State

Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China.

b. Xi'an High-tech Research Institute, 710025 Xi'an, China.

c. Xi'an Fiber Textile Supervision and Inspection Institute, Xi'an 710068, China.

*Corresponding authors: E-mail: zhaomshu@xjtu.edu.cn;
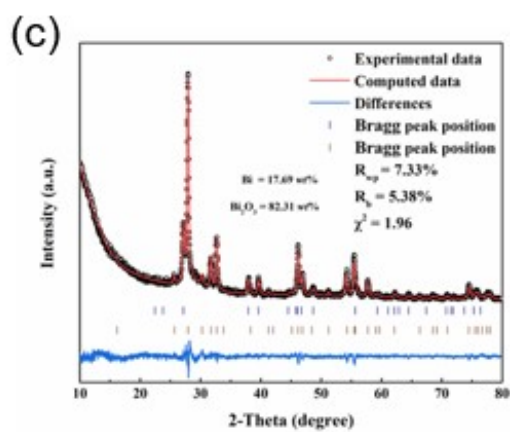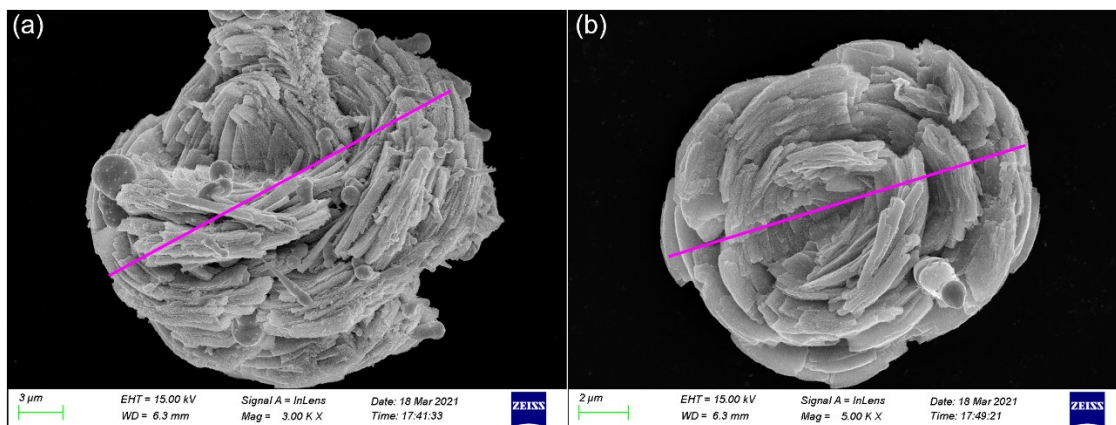
Tel: +86-13186193932

Fig. S1. (a, b)Low-magnification SEM images of Bi-Bi$_2$O$_3$ and (c) the Rietveld refinements on the
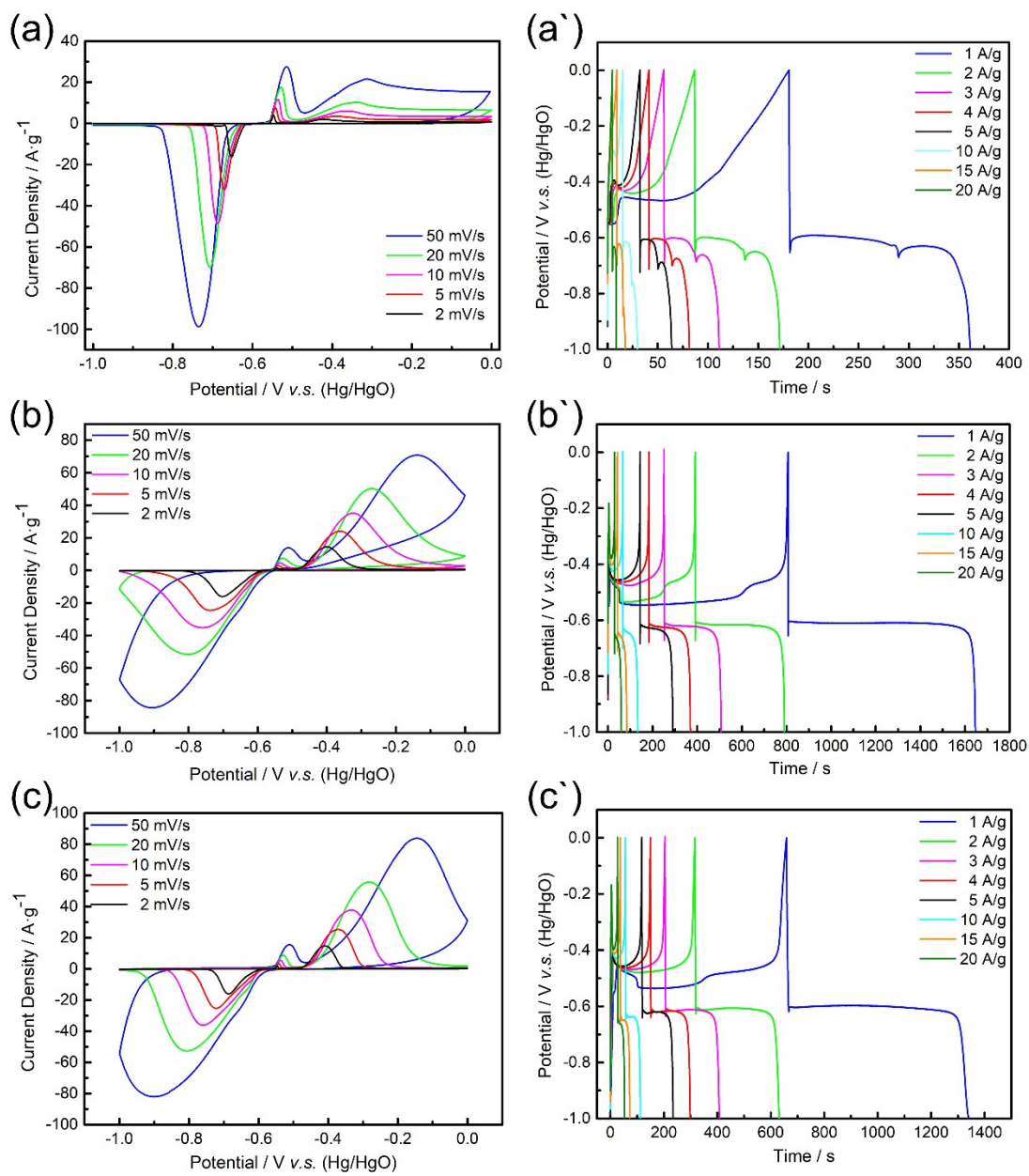
XRD pattern of the Bi-Bi$_2$O$_3$/rGO composites.

Fig. S2. (a-c) CV curves at different scan rates and (a`-c`) the corresponding GCD curves at various current densities of Bi-precursor, Bi-Bi$_2$O$_3$ and Bi-Bi$_2$O$_3$/rGO-60.

Table S1. The parameter information of the as-obtained electrosynthesis samples.

Sample	$Co(NO_3)_2 \cdot 6H_2O$ (mmol)	$Ni(NO_3)_2 \cdot 6H_2O$ (mmol)	Chronopotentiometry
$Ni(OH)_2$	None	6	
Ni_2Co_1-LDH	4	2	
NiCo-LDH	3	3	-1 V *vs.* Ag/AgCl for 200 s
Ni_1Co_2-LDH	2	4	
$Co(OH)_2$	6	None	

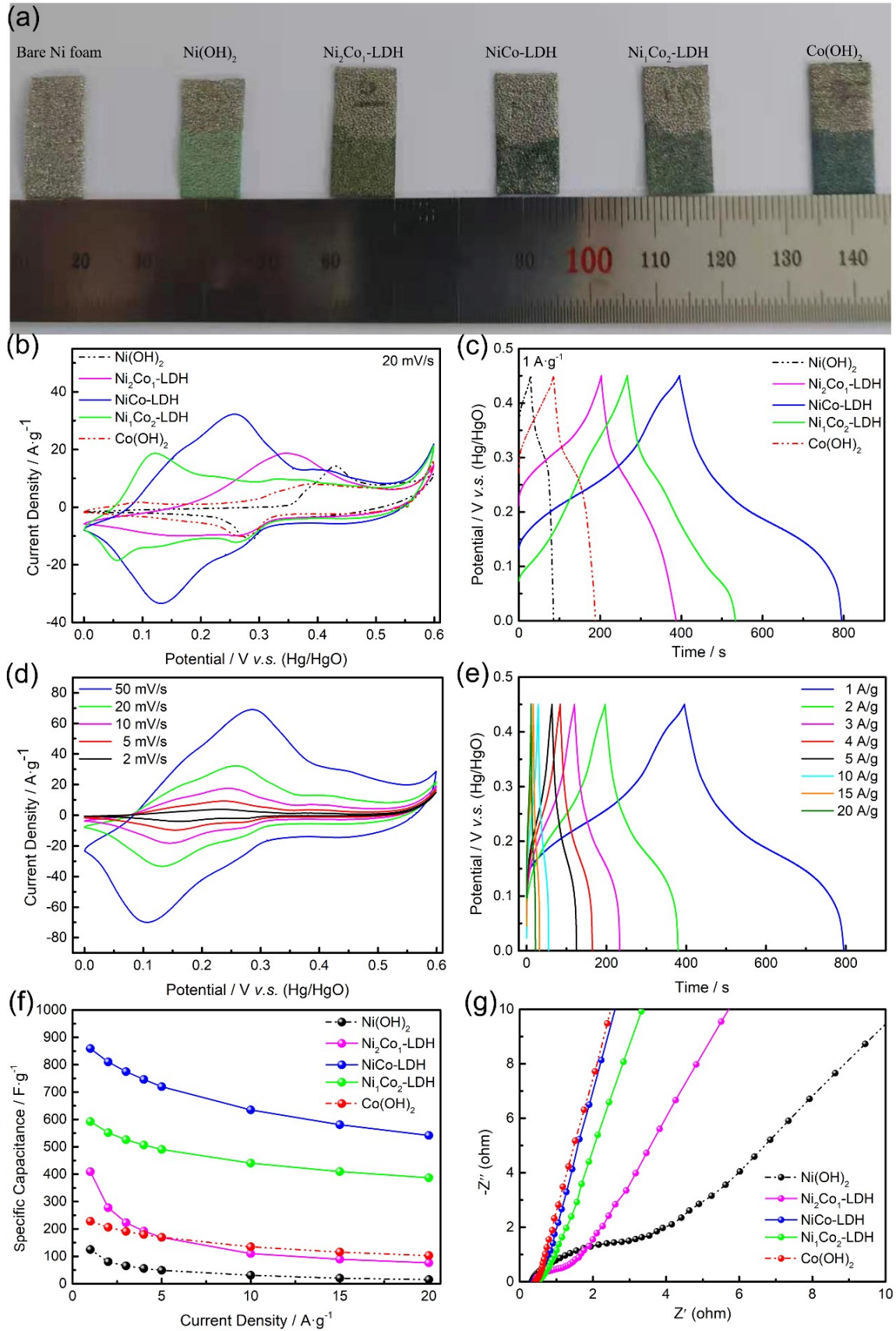| Sample	$Co(NO_3)_2 \cdot 6H_2O$ (mmol)	$Ni(NO_3)_2 \cdot 6H_2O$ (mmol)	Chronopotentiometry
$Ni(OH)_2$			
Ni_2Co_1-LDH			

Fig. S3. Optical images and a comparison of the electrochemical performance of the Ni(OH)$_2$, Ni$_2$Co$_1$-

LDH, NiCo-LDH, Ni_1Co_2-LDH and $Co(OH)_2$ electrodes in three-electrode system. (a) Optical images of the samples, (b) CV curves at a scan rate of 20 mV s^{-1}, (c) GCD curves at a current density of 1 A g^{-1}, (d, e) CV curves at various scan rate and GCD curves at different current densities of the NiCo-LDH electrode, (f) Specific capacitance at various current densities and (g) Nyquist plots of EIS.

Table S2. Impedance parameters simulated from the equivalent circuits.

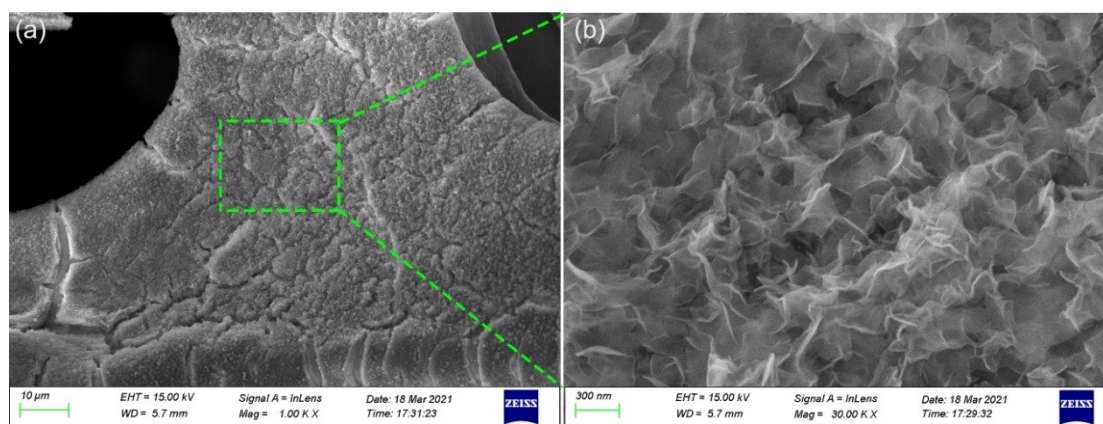| Sample	R_s (Ω)	R_{ct} (Ω)
$Ni(OH)_2$	0.32	3.49
Ni_2Co_1-LDH	0.39	0.35
NiCo-LDH	0.37	0.09
Ni_1Co_2-LDH	0.45	0.14
$Co(OH)_2$	0.35	---

Fig. S4. Low and high-magnification SEM images of NiCo-LDH via an electrosynthesis method.
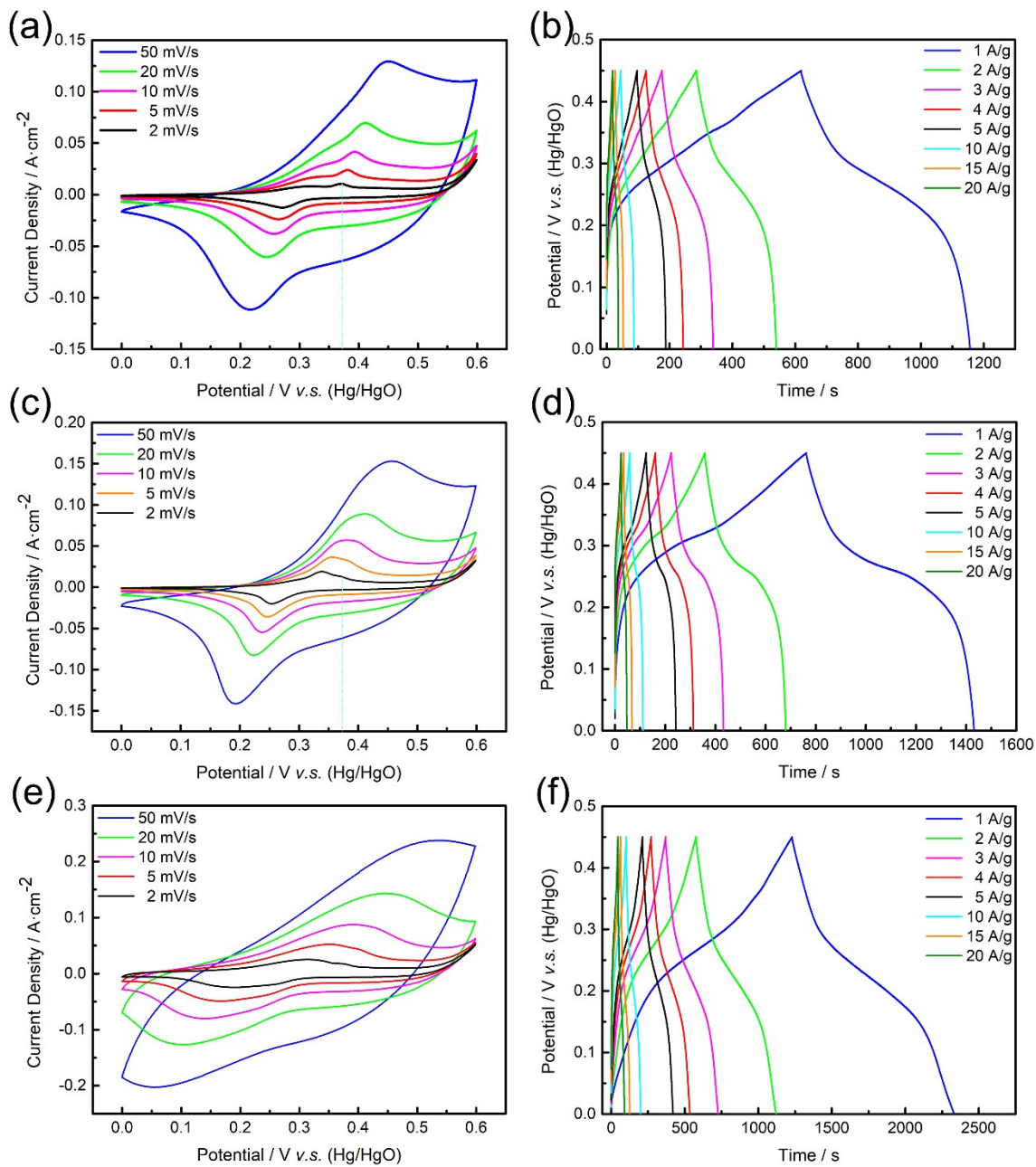
Fig. S5. CV curves at various scan rates and GCD curves at different current densities of (a, b) CoS_x, (c, d) $CoVS_x$ and (e, f) $CoVS_x$@NiCo-LDH.

Table S3. A comparison of electrochemical performance with previously reported work.

Material	Surface morphology	Capacitance	Current density	Electrolyte	Ref.
Bi_2O_3/rGO-20	Nanoparticles/Nanosheet	288.0 mAh g^{-1} (1036.9 F g^{-1})	1 A g^{-1}	6 M KOH	This work
HHP Bi_2O_3	Hollow hexagonal prism	327 mAh g^{-1}	1 A g^{-1}	6 M KOH	[1]
Bi_2O_2Se/rGO	Nanosheet/Nanosheet	258.11 mAh g^{-1}	1 A g^{-1}	1 M KOH	[2]
Bi_2O_3	Nanowires	691.3 F g^{-1}	2 A g^{-1}	6 M KOH	[3]
Bi_2O_3/NCDs	Unique structure	1046 F g^{-1}	1 A g^{-1}	3 M KOH	[4]
Bi-Bi_2O_3/CNT	Nanoparticles/Nanowires	850 F g^{-1}	1 A g-1	6 M KOH	[5]
Bi_2O_3/NF	Nanosheet-like	138.3 mA h g^{-1}	10 mA cm^{-2}	3 M KOH	[6]
Bi-Bi_2O_3	Nanosheets	252.5 mAh g^{-1}	2 A g^{-1}	0.5 M Na_2SO_4	[7]
(Ni,Co)Se_2/NiCo-LDH	Core/Shell Nanospheres	170 mAh g^{-1}	2 A g^{-1}	3 M KOH	[8]
Ni_3S_2@Ni(OH)$_2$-G	Sandwich	2258 F g^{-1}	1 A g^{-1}	3 M KOH	[9]
NiCo-LDH	Thiourea additive	1198 F^{-1}	1 A g^{-1}	2 M KOH	[10]
CC/NiCoP@NiCo-LDH	Core/shell heterostructure	1951 F g^{-1}	1 mA cm^{-2}	6 M KOH	[11]
CoVS$_x$@NiCo-LDH	Nanosheet/Nanosheet	306.0 mAh g^{-1} 2448.0 F g^{-1}	1 A g^{-1}	6 M KOH	This work

[1]   G. Zan, T. Wu, P. Hu, Y. Zhou, S. Zhao, S. Xu, J. Chen, Y. Cui, Q. Wu, An approaching-theoretical-capacity anode material for aqueous battery: Hollow hexagonal prism Bi_2O_3 assembled by nanoparticles, Energy Storage Mater. 28 (2020) 82-90. doi:10.1016/j.ensm.2020.02.027.

[2]   Y. Liu, M. Li, G. Wang, L. Dang, F. Li, M.E. Pam, H. Zhang, J. Han, H.Y. Yang, Bismuth Oxide Selenium/Graphene Oxide Composites: Toward High-Performance Electrodes for Aqueous Alkaline Battery, Energy Environ. Mater. (2020) 1-9. doi:10.1002/eem2.12130.

[3]   Y. Qiu, H. Fan, X. Chang, H. Dang, Q. Luo, Z. Cheng, Novel ultrathin Bi_2O_3 nanowires for supercapacitor electrode materials with high performance, Appl. Surf. Sci. 434 (2018) 16-20.

doi:10.1016/j.apsusc.2017.10.171.

[4] Z. Ji, W. Dai, S. Zhang, G. Wang, X. Shen, K. Liu, G. Zhu, L. Kong, J. Zhu, Bismuth oxide/nitrogen-doped carbon dots hollow and porous hierarchitectures for high-performance asymmetric supercapacitors, Adv. Powder Technol. 31 (2020) 632-638. doi:10.1016/j.apt.2019.11.018.

[5] H. Wu, J. Guo, D. Yang, Facile autoreduction synthesis of core-shell Bi-Bi2O3/CNT with 3-dimensional neural network structure for high-rate performance supercapacitor, J. Mater. Sci. Technol. 47 (2020) 169-176. doi:10.1016/j.jmst.2020.02.007.

[6] Y. Huang, L. Quan, T. Liu, Q. Chen, D. Cai, H. Zhan, Construction of MOF-derived hollow Ni-Zn-Co-S nanosword arrays as binder-free electrodes for asymmetric supercapacitors with high energy density, Nanoscale. 10 (2018) 14171-14181. doi:10.1039/c8nr03919d.

[7] T. Qin, D. Wang, X. Zhang, Y. Wang, N.E. Drewett, W. Zhang, T. Dong, T. Li, Z. Wang, T. Deng, Z. Pan, N. Yue, R. Yang, K. Huang, S. Feng, R. Huang, W. Zheng, Unlocking the Optimal Aqueous δ-Bi2O3 Anode via Unifying Octahedrally Liberated Bi-Atoms and Spilled Nano-Bi Exsolution, Energy Storage Mater. 36 (2021) 376-386. doi:10.1016/j.ensm.2021.01.013.

[8] X. Li, H. Wu, C. Guan, A.M. Elshahawy, Y. Dong, S.J. Pennycook, J. Wang, $(Ni,Co)Se_2$/NiCo-LDH Core/Shell Structural Electrode with the Cactus-Like $(Ni,Co)Se_2$ Core for Asymmetric Supercapacitors, Small. 15 (2019) 1-10. doi:10.1002/smll.201803895.

[9] X. Wang, J. Hu, Y. Su, J. Hao, F. Liu, S. Han, J. An, J. Lian, Ni Foam-Ni_3S_2@$Ni(OH)_2$-Graphene Sandwich Structure Electrode Materials: Facile Synthesis and High Supercapacitor Performance, Chem.-A Eur. J. 23 (2017) 4128-4136. doi:10.1002/chem.201605212.

[10] Y. Wang, Z. Yin, G. Yan, Z. Wang, X. Li, H. Guo, J. Wang, New insight into the

electrodeposition of NiCo layered double hydroxide and its capacitive evaluation, Electrochim. Acta. 336 (2020) 135734. doi:10.1016/j.electacta.2020.135734.

[11]   X. Gao, Y. Zhao, K. Dai, J. Wang, B. Zhang, X. Shen, NiCoP nanowire@NiCo-layered double hydroxides nanosheet heterostructure for flexible asymmetric supercapacitors, Chem. Eng. J. 384 (2020) 123373. doi:10.1016/j.cej.2019.123373.