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Electronic Supplementary Information (ESI)

Tailoring the morphology followed electrochemical performance of NiMn-LDH

nanosheet-arrays through controlled Co-doping for high-energy and power

asymmetric supercapacitors

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Fig. \$1 (a) XRD patterns of NiCoMn-LDH (10%) on the Ni-foam substrate.



Fig. \$2 The XPS spectra showing relative shifts in binding energy values of the constituent elements i.e. (a) Ni2p, (b) Mn2p, (c) Co2p, and (d) O1s for undoped and Co-doped NiMn-LDH samples.



Fig. \$3 FE-SEM images of different Co-doped samples as (a, a') NiMn-LDH; (5%), (b, b') NiCoMn-LDH (15%) and (c, c') NiCoMn-LDH (20%).



Fig. \$4 N2 adsorption/desorption isotherms and BJH pore size distribution curve of Co-doped NiMn-LDH samples for different doping percentage.



Fig. \$5. Cycling stability study of Co-doped NiMn-LDH samples for different doping percentage

Fabrication of NiCoMn-LDH (10%)//RGO ASC device

Firstly, RGO for negative electrode has been obtained same as in our previous reported work¹. Further, for the construction of negative electrode active material has been mixed with acetylene black and polytetrafluoroethylene (PTFE) suspension (60 wt. %) binder at a weight ratio of 8:1:1, and then pressed onto a nickel foam (20 MPa), which served as a current collector. Finally, we dried it in a vacuum oven at 60 °C for 12 h.

Positive electrode has been used as it is, obtained by the hydrothermal deposition of NiCoMn-LDH (10%) material on the Ni foam. Afterwards, both of these two electrodes have been assembled by electrically isolating each other using separator (Celguard-3501 porous membrane). Importantly, KOH electrolyte of molarity 3 M is used for filling the cylindrical tube. From the FE-SEM images, randomly aligned RGO NS aggregate with each other can be clearly seen (Fig \$6(a)). To investigate the capacitive behavior, CV and GCD measurements of RGO were also performed (Fig. \$6(b, c). Fig. \$6(b) shows the CV curves of RGO at different scan rates (5-50 mV.s⁻¹). The rectangular shape of CV profile is observed, indicating electrical double layer behavior of RGO. The calculated SC value of RGO electrode from its GCD has been reached to 184 F. g⁻¹ at 1 A. g⁻¹ in the voltage range of -1.0 to 0.0 V using formula $SC = \frac{I\Delta t}{m\Delta V}$, where, I is the discharge current, Δt is discharge time, m is a mass of the RGO material about 3.mg. Furthermore, this performance still shows high SC value 100 F. g⁻¹, as current density is increased to 15 A. g⁻¹ (Fig. \$6d).



Fig. \$6 (a) FE-SEM image of RGO sample, (b) CV and (c) GCD curves of RGO at different scan rates and at different current densities and (d) SC of RGO electrode at different current densities.

Reference

1. Q. Xia, K. Hui, K. Hui, S. Kim, J. Lim, Si. Choi, L. Zhang, R. Mane, J. Yun, and K. H. Kim; *J. Mater. Chem.*, A 2015, **3**, 22102-22117

Sample	<i>d</i> ₀₀₃	<i>d</i> ₀₀₆	<i>d</i> ₁₁₀	c (Å)	a (Å)	<i>d</i> (nm)	Micro
	(Å)	(Å)	(Å)			(003)	strain
							(E ₀₀₃)
NiMn-LDH (0%)	7.95	4.00	1.555	23.85	3.11	270	0.052937
NiCoMn-LDH (5%)	7.96	3.99	1.550	23.88	3.10	289	0.0494791
NiCoMn-LDH (10%)	7.96	3.99	1.545	23.88	3.10	289	0.0494431
NiCoMn-LDH (15%)	7.97	4.00	1.540	23.91	3.08	289	0.0499084
NiCoMn-LDH (20%)	8.00	4.00	1.530	24.00	3.06	308	0.0460093

 Table \$1: Effect of Co-doping levels on structural parameters of NiMn-LDH.

Table \$2: Effect of Co-doping levels on surface-area, pore-diameter and pore-volume of NiMn-LDH (obtained from Fig. \$4).

Sample	Surface-area	Pore-diameter	Pore-volume
	$(m^2 g^{-1})$	(nm)	$(cm^3 g^{-1})$
NiMn-LDH (0%)	82	17	0.93
NiCoMn-LDH (5%)	94	14	0.97
NiCoMn-LDH (10%)	102	13	1.03
NiCoMn-LDH (15%)	78	10	0.81
NiCoMn-LDH (20%)	73	9	0.71