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Supplementary Information

Core/Shell Design of Efficient Electrocatalysts Based on $NiCo_2O_4$ Nanowires and NiMn LDH Nanosheets for Rechargeable Zinc-Air Batteries

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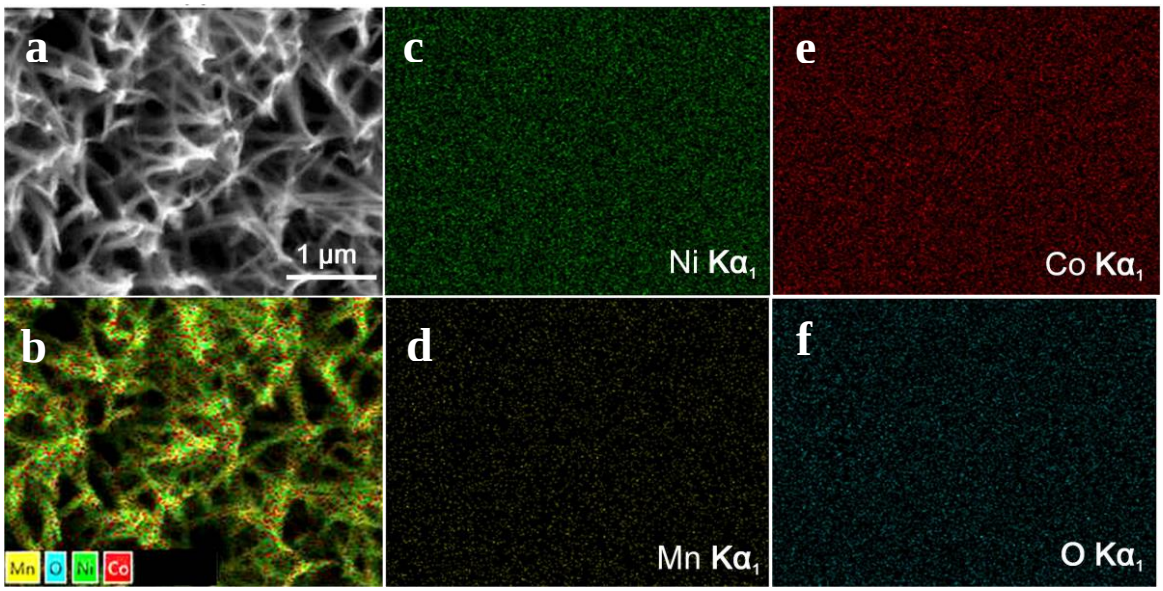

Fig. S1 EDS mapping of $NiCo_2O_4$@NiMn LDH core/shell arrays. (a) SEM images, (b) Hierarchical electronic image, (c-f) Distributions of Ni, Mn, Co and O elements.

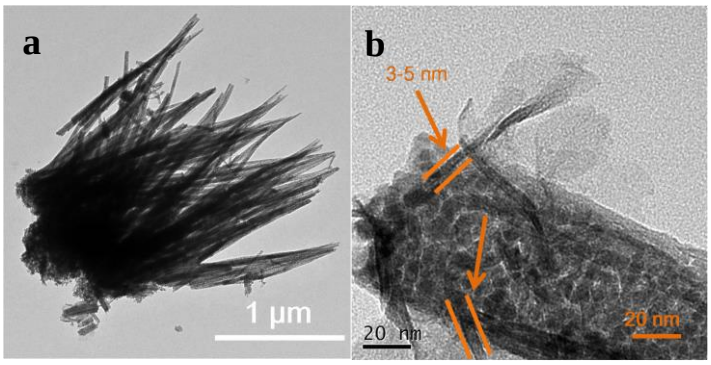

Fig. S2 (a) Integrated $NiCo_2O_4$ nanowires. (b) TEM image of $NiCo_2O_4$@NiMn LDH.

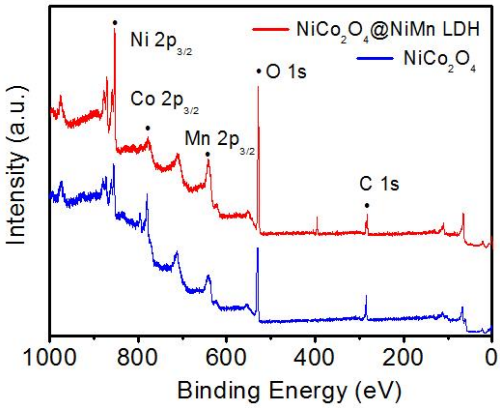

Fig. S3 The survey XPS spectrums for $NiCo_2O_4$ and $NiCo_2O_4$@NiMn LDH.

Table S1 The electrocatalytic activities of the recently reported NiCo oxides and LDHs electrocatalysts for OER.

Materials	Electrolyte	Loading (mg cm^{-2})	Overpotential (mV vs. RHE) at 10 mA cm^{-2}	Onset potential (mV vs. RHE)	References
$NiCo_2O_4$@NiMn LDH arrays@Ni foam	**1.0 M KOH**	**2.0**	**255**	**216**	**This work**
NiCo oxides catalysts					
$Ni_xCo_{3-x}O_4$ nanosheets @Ni foil	0.1 M KOH	0.127	530	340	**Chem. Commun.** 2015, 51, 9511. (S1)
$NiCo_2O_4$/N-doped-graphene paper	0.1 M KOH	----	434	310	**ACS Nano** 2013, 7, 10190. (S2)
$NiCo_2O_4$/CNTs*	1.0 M KOH	0.2	420	240	**J. Mater. Chem. A** 2015, 3, 19314. (S3)
$NiCo_2O_4$ nanowires*	0.1 M KOH	----	412	337	**J. Mater. Chem. A** 2013, 1, 12170. (S4)
$Ni_xCo_yO_4$/Co-N-rGO*	0.1 M KOH	0.2	400	230	**J. Mater. Chem. A** 2017, 5, 5594. (S5)
$NiCo_2O_4$/graphene /MnO_2@Ni foam	0.1 M KOH	12.8	371	321	**Chem. Commun.** 2014, 50, 207. (S6)
$Ni_xCo_{3-x}O_4$ nanowire arrays@Ti foils	1 M NaOH	~3.0	370	----	**Adv. Mater.** 22 (2010) 1926. (S7)
NiCo/$NiCoO_x$ nanowire arrays@Ni foam	1.0 M KOH	0.7	361	270	**ACS Appl. Mater. Inter.** 2016, 8, 3208. (S8)
$NiCo_2O_4$ nanowires @Ti foils	1.0 M KOH	0.3	350	330	**Adv. Energy Mater.** 2015, 5, 1402031. (S9)
CoO/N-doped graphene*	1.0 M KOH	0.7	340	----	**Energy Environ. Sci.** 2014, 7, 609. (S10)
Spinel Ni-Co oxide array @FTO	1.0 M NaOH	----	340	270	**Adv. Energy Mater.** 2015, 5, 1500091. (S11)
Au/ $NiCo_2O_4$ array @Ti foil	1.0 M KOH	0.5	330	240	**ChemCatChem** 2014, 6, 2501. (S12)
$NiCo_2O_4$/$NiCo_2O_4$ arrays @Carbon cloth	1.0 M NaOH	----	320	280	**Nano Energy** 2015, 11, 333. (S13)
$Ni_{0.88}Co_{1.22}Se_4$ Microparticles@ FTO	1.0 M KOH	0.096	320	270	**Chem. Mater.** 2017, 29, 7032. (S14)
$NiCo_2O_4$ nanosheets with oxygen vacancies*	1.0 M KOH	0.285	320	300	**Angew. Chem. Int. Ed.** 2015, 127, 7507. (S15)
Co_3O_4/N-doped-graphene @Ni foam	1.0 M KOH	1.0	310	270	**Nat. Mater.** 2011, 10, 780. (S16)
$NiCo_2N$ nanosheet attays @Ni foam	1.0 M KOH	----	290	260	**ChemSusChem** 2017, 10, 4170. (S17)

$NiCo_2O_4$ hollow microcuboids@Ni foam	1.0 M NaOH	1.0	290	230	**Angew. Chem. Int. Ed.** 2016, 55, 6290. (S18)
$NiCo_2O_4$/NiCoB array@Carbon cloth	1.0 M KOH	2.3	270	----	**Inorg. Chem. Front.** 2017, 4, 1546. (S19)
NCNT/CoO/NiO/NiCo*	1.0 M KOH	0.21	270	----	**Angew. Chem. Int. Ed.** 2015, 54, 9654. (S20)
1D $LaCo_{1-x}Ni_xO_{3-\delta}$*	1.0 M KOH	0.7	265	----	**ACS Appl. Mater. Inter.** 2016, 8, 6019. (S21)
LDHs catalysts					
ZnCo LDH array@Ni foil	0.1 M KOH	----	512	330	**J. Mater. Chem. A** 2014, 2, 13250. (S22)
NiCo LDH nanosheets@ Ni foam	0.1 M KOH	1.75	420	290	**J. Power Sources** 2015, 278, 445. (S23)
Unilamellar NiCo LDH @Carbon paper	1.0 M KOH	0.17	367	290	**Nano Lett.** 2015, 15, 1421. (S24)
NiMn LDH nanoflowers*	1.0 M KOH	0.2	350	260	**Nanoscale** 2017, 9, 774. (S25)
Unilamellar CoCo LDH*	1.0 M KOH	0.07	335	393	**Nat. Commun.** 2014, 5, 4477. (S26)
CoMn LDH nanosheets@ Carbon fiber paper	1.0 M KOH	0.142	324	265	**J. Am. Chem. Soc.** 2014, 136, 16481. (S27)
NiFe-LDH/Co,N- carbon nanoframes*	0.1 M KOH	0.12	312	270	**Adv. Energy Mater.** 2017, 7, 1700467. (S28)
Unilamellar NiFe LDH*	1.0 M KOH	0.07	300	250	**Nat. Commun.** 2014, 5, 4477. (S26)
NiFeMn LDH @Carbon fiber paper	1.0 M KOH	0.2	265	200	**Chem. Commun.** 2016, 52, 908. (S29)
Unilamellar NiMn LDH nanosheets/rGO*	1.0 M KOH	0.2	260	190	**Nanoscale** 2016, 8, 10425. (S30)
NiFe LDH/CNT @Carbon fiber paper	1.0 M KOH	0.25	247	220	**J. Am. Chem. Soc.** 2013, 135, 8452. (S31)
NiFe LDH arrays @Ni foam	1.0 M KOH	----	244	220	**Nat. Commun.** 2016, 7, 12324. (S32)
Commercial IrO_2@Ni foam	1.0 M KOH	0.7	285	220	**ACS Appl. Mater. Inter.**2016, 8, 3208. (S8)
Commercial Ir/C@ Carbon fiber paper	1.0 M KOH	0.2	~280	220	**J. Am. Chem. Soc.** 2013, 135, 8452. (S31)

* Denoted that the work substrates of those electrocatalysts were glassy carbon electrode.

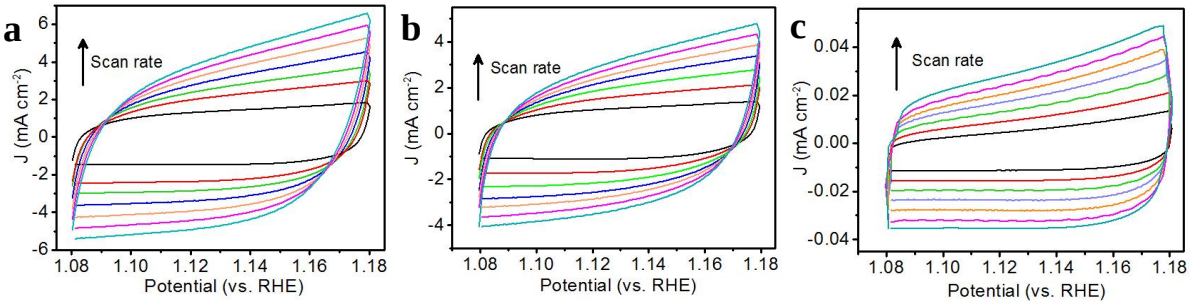

Fig. S4 Cyclic voltammograms in the double layer region at scan rates of 15, 13, 11, 9, 7, 5, 3 mV s^{-1} (along the arrow direction) of (a) $NiCo_2O_4$@NiMn LDH core/shell arrays, (b) $NiCo_2O_4$ nanowires and (c) NiMn LDH.

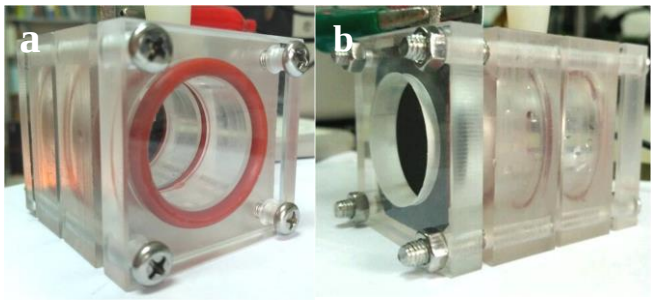

Fig. S5 Photographs of liquid Zn-air battery.

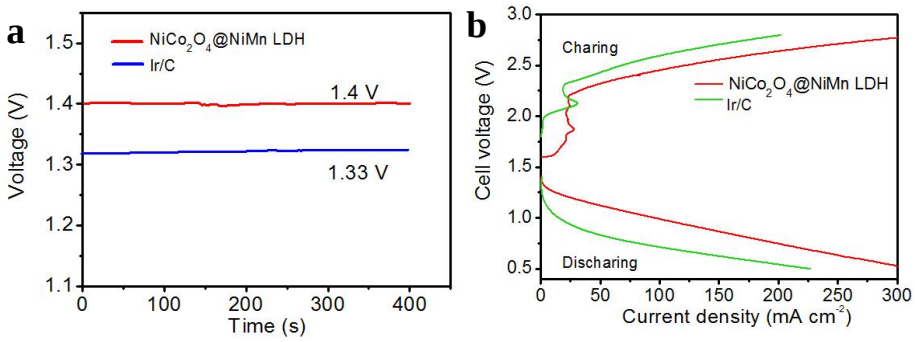

Fig. S6 (a) Charge and discharge polarization curves of the rechargeable Zn-air batteries with various electrocatalysts. (b) Open-circuit curves of the rechargeable Zn-air batteries with various electrocatalysts.

Movie S1. The $NiCo_2O_4$@NiMn LDH electrode operated at a current density of 150 mA/cm^2 to drive OER.

Table S2 The performance of the primary Zn-air batteries with various electrocatalysts.

Electrocatalysts	Loading (mg cm^{-2})	Peak power density (mW cm^{-2})	Specific capacity (mA h g_{Zn}^{-1})	Energy density (Wh kg_{Zn}^{-1})	References
$NiCo_2O_4$/NiMn LDH arrays@Ni foam	**2.0**	**160.8**	**722**	**866**	**This work**
MnO_x@stainless steel	0.3	26	----	----	**Energy Environ. Sci.** 2014, 7, 2017. (S33)
Co_3O_4 nanowires @stainless steel mesh	1.5	40	----	----	**Adv. Energy Mater.** 2014, 4, 1301389. (S34)
$Fe_{0.1}Ni_{0.9}Co_2O_4$/Ti and MnO_2/carbon black	----	88.8	----	----	**J. Power Sources** 2016, 313, 37. (S35)
Fe-N_x/N/S-doped carbon	1.25	102.7	----	----	**Angew. Chem. Int. Ed.** 2017, 56, 610. (S36)
Mn_3O_4/graphene	----	120	----	----	**Energy Environ. Sci.** 2011, 4, 4148. (S37)
$NiCo_2S_4$/N-CNT	1.0	147	431.1	554.6	**Nano Energy** 2017, 31, 541. (S38)
Porous N-doped graphene	----	70	400	460	**J. Mater. Chem.** 2012, 22, 12810. (S39)
$ZnCo_2O_4$ quantum dots /NCNT	2.0	82.3	428.47	595.57	**Adv. Mater.** 2016, 28, 3777. (S40)
Ni_3Fe/N-doped carbon sheets	----	----	528	634	**Adv. Energy Mater.** 2017, 7, 1601172. (S41)
c-$CoMn_2$/C	2.0	79	~500	650	**Nat. Commun.** 2015, 6, 7345. (S42)
N-CNTs/$Co_xMn_{1-x}O$	0.53	----	581	695	**Nano Energy** 2016, 20, 315. (S43)
N-doped CNT/ CoO-NiO-NiCo	0.53	----	594	713	**Angew. Chem. Int. Ed.** 2015, 54, 9654. (S20)
1D $NiCo_2O_4$	----	----	580	725	**Nanoscale** 2014, 6, 3173. (S44)
Cu-Pt nanocage	2.0	250	560	728	**ACS Catal.** 2015, 5, 1445. (S45)
NiFe nanoparticles /N-Graphene		~85	583.7	732.3	**ACS Catal.** 2016, 6, 6335.(S46)
N-doped-CNTs	----	190	623.4	776.5	**Nano Energy** 2017, 37, 98. (S47)
$NiCo_2O_4$/C@Ni foam	2.3	----	688	826	**Nano Lett.** 2017, 17, 156. (S48)
N,P-doped carbon foam	0.5	55	735	835	**Nat. Nanotech.** 2015, 10, 444. (S49)
NiO/CoN Porous nanowires	----	79.6	648	836	**ACS Nano** 2017, 11, 2275. (S50)

N-doped carbon fiber film	2.0	185	660	838	**Adv. Mater.** 2016, 28, 3000. (S51)
Co_4N/N-C fibers	----	174	774	975	**J. Am. Chem. Soc.** 2016, 138, 10226. (52)

Table S3 The performance of the rechargeable Zn-air batteries with various electrocatalysts.

Electrocatalysts	Loading (mg cm^{-2})	Initial round-trip efficiency	Cycling conditions	Increasing of voltage gap	References
$NiCo_2O_4$/NiMn LDH arrays@Ni foam	**2.0**	**63%** **56%**	**500 cycles (10 min/cycle, 5 mA cm^{-2})** **500 cycles (10 min/cycle, 20 mA cm^{-2})**	**0.045 V** **0.02 V**	**This work**
1D $NiCo_2O_4$	----	61%	50 cycles (20 min/cycle, 20 mA cm^{-2})	0.14 V	**Nanoscale** 2014, 6, 3173. (S44)
NiMn LDH nanofolwers	1.0	~58%	200 cycles (10 min/cycle, 10 mA cm^{-2})	~0.45 V	**Nanoscale** 2017, 9, 774. (S25)
$Pb_2Ru_2O_{6.5}$nanoparticles	---	~61%	18 cycles (2 h /cycle, 20 mA cm^{-2})	~0.36	**Energy Environ. Sci.** 2017,10,129. (S53)
$CuCo_2O_4$@CNTs	1.0	~58%	60 cycles (10 min/cycle, 10 mA cm^{-2})	~0.22 V	**Nano Lett.** 2017, 17, 7989. (S54)
Co_3O_4 nanowires @stainless steel mesh	1.5	~40%	100 cycles (10 min/cycle, 17.6 mA cm^{-2})	~0.1 V	**Adv. Energy Mater.** 2014, 4, 1301389. (S34)
$Co(OH)_2$+N-rGO	1.0	46%	75 cycles (40 min/cycle, 15 mA cm^{-2})	0.2 V	**ACS Appl. Mater.** Inter. 2015, 7, 12930. (S55)
$LaNiO_3$/N-CNT	0.72	41%	75 cycles (10 min/cycle, 17.6 mA cm^{-2})	0.15 V	**Nano Lett.** 2012, 12, 1946. (S56)
NiO/CoN Porous Nanowires	----	61.6%	50 cycles (10 min/cycle, (50 and 1 mA cm^{-2})	~0.06	**ACS Nano** 2017, 11, 2275. (S50)
B/N co-doped porous carbon	3	46%	66 cycles (10 min/cycle, 20 mA cm^{-2})	0.02 V	**Adv. Mater.** 2015, 27, 3789. (S57)
CoO/N-CNT and	1.0+5.0	63%	60 cycles	negligible change	**Nat. Commun.**

NiFe LDH/Ni foam			(200 s/cycle, 10 mA cm^{-2})		2013, 4, 1805. (S58)
P-doped-g-C_3N_4@ Carbon-fiber paper	----	42.6%	50 cycles (20 min/cycle, 20 mA cm^{-2})	negligible change	**Angew. Chem. Int. Ed.** 2015, 54, 4646. (S59)
N,P-doped carbon foam	0.5	59%	180 cycles (10 min/cycle, 2 mA cm^{-2})	0.7 V	**Nat. Nanotechnol.** 2015, 10, 444. (S49)
$La_x(Ba_{0.5}Sr_{0.5})_{1-x}Co_{0.8}Fe_{0.2}O_{3-\delta}$	0.64	62.5%	100 cycles (10 min/cycle, (20 mA cm^{-2})	0.25 V	**Energy Environ. Sci.** 2016, 9, 176. (S60)
Fe nanoparticles/ N-doped carbon nanoshell	2.2	64%	100 cycles (10 min/cycle, 10 mA cm^{-2})	0.16 V	**Nano Energy** 2015, 13, 387. (S61)
Fe-N_x@N-S/carbon layers@CNTs	1.25	~60%	100 cycles, (5 mA cm^{-2})	~0.15 V	**Angew. Chem. Int. Ed.** 2017, 56, 610. (S36)
Fe_3Mo_3/C@IrMn nanoalloys	10.0	61%	100 cycles (1 h/cycle, 10 mA cm^{-2})	0.11 V	**Adv. Mater.** 2017, 29, 1702385. (S62)
$NiCo_2S_4$/N-CNT	2.0	67.2%	150 cycles (400 s/cycle, (10 mA cm^{-2})	0.06 V	**Nano Energy** 2017, 31, 541. (S38)
c-$CoMn_2$/C	2.0	----	155 cycles (400 s/cycle, (10 mA cm^{-2})	0.06 V	**Nat. Commun.** 2015, 6, 7345. (S42)
$Y_2[Ru_{2-x}Y_x]O_{7-y}$ nanocrystalline	----	61%	200 cycles (10 min/cycle, 10 mA cm^{-2})	0.1 V	**Nano Lett.** 2017, 17, 3974. (S63)
NiCo nanoparticles @Fibrous carbon aerogels	1.0	65.4%	300 cycles (2 h /cycle, 10 mA cm^{-2})	0.2 V	**Nano Lett.** 2016, 16, 6516. (S64)
N-doped carbon fiber film	2.0	62%	500 cycles (20 min/cycle, 10 mA cm^{-2})	0.13 V	**Adv. Mater.** 2016, 28, 3000. (S51)

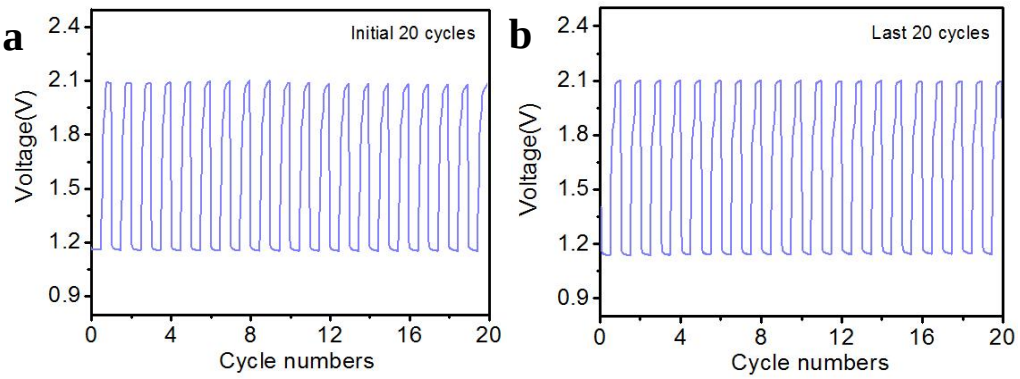

Fig. S7 The initial (a) and last (b) discharge / charge voltage profiles of Zn–air batteries with $NiCo_2O_4$@NiMn LDH.

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