## Supplementary Information

## **Regulation of activity interface in Ni-Co-Mn ternary aerogel catalysts**

## for efficient Water Splitting

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Figure S1. Optical photographs of the precursor solution and the resulting wet gel.



Figure S2. (a) SEM mapping of NCM111. (b) TEM image of NCM111.



Figure S3. (a) The HER overpotential of NCM111 at the current density of 20 mA/cm<sup>2</sup>. (b) The HER overpotential of NCM111 at the current density of 100 mA/cm<sup>2</sup>. (c) HER polarization curves of bare carbon aerogel and Ni foam in 1.0 M KOH at a scan rate of 5 mV/s.



Figure S4. (a) The OER overpotential of NCM111 at the current density of 20 mA/cm<sup>2</sup>. (b) The OER overpotential of NCM111 at the current density of 100 mA/cm<sup>2</sup>. (c) OER polarization curves of bare carbon aerogel and Ni foam in 1.0 M KOH at a scan rate of 5 mV/s.



Figure S5. (a) CVs of NCM111 at potential from 0.1 V to 0.3 V vs RHE at scan rates of 20 mV/s, 50 mV/s, 80 mV/s, 110 mV/s and 140 mV/s. (b) The capacitive current densities of NCM111 measured at 0.1 V vs RHE with different scan rates. (c) CVs of NCM523 at potential from 0.1 V to 0.3 V vs RHE at scan rates of 20 mV/s, 50 mV/s, 80 mV/s, 110 mV/s and 140 mV/s. (d) The capacitive current densities of NCM523 measured at 0.1 V vs RHE with different scan rates.



Figure S6. (a) CVs of NCM622 at potential from 0.1 V to 0.3 V vs RHE at scan rates of 20 mV/s, 50 mV/s, 80 mV/s, 110 mV/s and 140 mV/s. (b) The capacitive current densities of NCM622 measured at 0.1 V vs RHE with different scan rates. (c) CVs of NCM811 at potential from 0.1 V to 0.3 V vs RHE at scan rates of 20 mV/s, 50 mV/s, 80 mV/s, 110 mV/s and 140 mV/s. (d) The capacitive current densities of NCM811 measured at 0.1 V vs RHE with different scan rates.



Figure S7 SEM image of NCM111 after the stability evaluation.



Figure S8. Schematic diagram of molecular structure of HER process.



Figure S9. Schematic diagram of molecular structure of OER process.

Materials	HER	OER	Ref.
CuO–ZnO	low onset potential of 1.4 V	low onset potential of 0.15 V	[1]
CoP-InNC@CNT	10 mA cm <sup>2</sup> —159mV	10 mA cm <sup>2</sup> —270mV	[2]
Pt/C	10 mA cm <sup>2</sup> —37mV	/	
Ir/C	/	10 mA cm <sup>2</sup> —295mV	
MOF with Co <sub>2</sub> N <sub>2</sub> O <sub>8</sub> SBUs	10 mA cm <sup>2</sup> —196mV	10 mA cm <sup>2</sup> —180mV	[3]
Co/Ni-doped MAF-4/ZIF-8	10 mA cm <sup>2</sup> —183mV	10 mA cm <sup>2</sup> —313mV	[4]
RuO <sub>2</sub>	/	10 mA cm <sup>2</sup> —376mV	
IrO <sub>2</sub>	/	10 mA cm <sup>2</sup> —406mV	
Zn-doped RuO <sub>2</sub>	/	10 mA cm <sup>2</sup> —173mV	[5]
RuO <sub>2</sub>	/	100 mA cm <sup>2</sup> —377mV	
TiMn <sub>2</sub>	10 mA cm <sup>2</sup> —139mV	/	[6]
Pt/C	10 mA cm <sup>2</sup> —34mV	/	
Pt@RuO <sub>x</sub>	10 mA cm <sup>2</sup> —20mV	10 mA cm <sup>2</sup> —157mV	[7]
Tantalum-induced reconstruction of $Ni_3S_2$	10 mA cm <sup>2</sup> —56mV	10 mA cm <sup>2</sup> —242mV	[8]
Mo/1T-MOS <sub>2</sub>	10 mA cm <sup>2</sup> —151mV	/	[9]
Pt/C	10 mA cm <sup>2</sup> —73mV	/	
Fe/W-Ni <sub>3</sub> S <sub>2</sub>	10 mA cm <sup>2</sup> —222mV	10 mA cm <sup>2</sup> —174mV	[10]
FeCoNiPtRu high- entropy alloy	10 mA cm <sup>2</sup> —104mV	10 mA cm <sup>2</sup> —331mV	[11]
RuP/CoNiP <sub>4</sub> O <sub>12</sub> //CC	10 mA cm <sup>2</sup> —27mV	10 mA cm <sup>2</sup> —227mV	[12]
RuO <sub>2</sub>	/	100 mA cm <sup>2</sup> —345mV	
Superaerophobic nickel-coated catalysts on Chinese rice paper	10 mA cm <sup>2</sup> —87mV	20 mA cm <sup>2</sup> —250mV	[13]
NCM	10 mA cm <sup>2</sup> —198mV	20 mA cm <sup>2</sup> —358mV	This work

Table S1 The state-of-the-art comparison table with previously reported catalysts

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