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SUPPORTING INFORMATION

Development of Efficient Electrocatalyst *via* Molecular Hybridization of NiMn Layered Double Hydroxide Nanosheets and Graphene

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Figure S1. Typical EDS results of Cl⁻-intercalating Ni_{2/3}Mn_{1/3} LDH (NiMn-Cl) nanoplatelets.



Figure S2. a) TG-DTA curves of NiMn-Cl LDH nanoplatelets in the range of room temperature to 1000 °C, b) XRD patterns of resulted samples obtained at different temperature (i) original, (ii) 130 °C, (iii) 600 °C, (iv) 1000 °C.

Figure S2 shows the TG-DTA curves and XRD patterns of the calcined products at different temperature. As displayed in Figure S2a, the first mass loss of 1.8% blow 60 °C was attributed to the evaporation of adsorbed water on the NiMn LDH surface. With the increase of temperature to 130 °C, a mass change of ~8% was ascribed to the removal of interlayer water. When the temperature was raised up to 1000 °C, a weight loss of 23.7% was recorded. The calcined product of NiMn LDHs was identified as NiO (PDF # 47-1049) and NiMn₂O₄ (PDF # 01-1110) as shown by XRD patterns in Figure S2b(iv). The chemical composition of assynthesized NiMn LDH nanoplatelets was estimated to be Ni_{2/3}Mn_{1/3}(OH)₂Cl_{1/3}·0.5H₂O. The metal composition is consistent with 2:1 molar ratio of Ni/Mn salts in the starting solution.



Figure S3. XRD patterns of natural graphite (i), graphene oxide (GO) (ii) and reduced graphene oxide (rGO) (iii).



Figure S4. Elemental mapping profiles on NiMn LDH nanosheets (NiMn-NS) and graphene oxide (GO) hybrid composite (NiMn-GO). C map is from graphene, O map is from both LDHs and GO, Ni- and Mn-maps are derived from LDH nanosheets.



Figure S5. *iR*-corrected linear sweep voltammetry curves for OER catalysis (a) and Tafel plots (b) of NiMn LDH nanoplatelets intercalated with DS⁻ (NiMn-DS) and Cl⁻ (NiMn-Cl).



Figure S6. Current density differences ($\Delta j = j_a - j_c$) plotted against scan rate for Clintercalating NiMn LDH nanoplatelets (NiMn-Cl), DS-intercalating NiMn LDH nanoplatelets (NiMn-DS) and NiMn LDH nanosheets (NiMn-NS).



Figure S7. Current density at 1.58 V vs RHE against ECSA ($2C_{dl}$) for different catalysts (inset, the corresponding magnified part for NiMn-Cl, NiMn-DS and NiMn-NS).



Figure S8. *iR*-corrected linear sweep voltammetry curves for HER catalysis (a) and Tafel plot (b) of the composites hybridizing NiMn LDH nanosheets and reduced graphene oxide (NiMn-rGO).



Figure S9. Water-splitting test cell driven by a 1.5 V AA battery (see supplementary movie for the demonstration of gas evolution).