Electronic Supplementary Information

Arising Synergetic and Antagonistic Effects in the Design of Ni- and Ru-Based Water Splitting Electrocatalysts

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Fig. S1 SEM images of (a) the pristine sodium ruthenate following heating at 900°C (scale = 1 μm) and (b) the hydrated layered protonic ruthenate following proton exchange with HCl 1M (scale = 1 μm). (c) XRD patterns of the final TMA-intercalated exfoliated material. (d) AFM analysis of the resulting ruthenate nanosheets evidencing the atomically thin structure of these materials.

Fig. S2 (a) SEM images (scale = 100 nm) of pristine Ru-based electrode following heat treatment at 250°C under argon. (b) XRD patterns for pristine Ru-based references heat treated under argon flow at an increasing 250°C to 400°C temperature range, and alternatively following treatment under air flow at 250°C.
**Fig. S3** (a) XRD patterns of individually synthesized Ni₃P at 250°C under Ar flow and (b) corresponding SEM images (scale = 100 nm).

**Fig. S4** EDX results evidencing the presence of Ru, Ni, and P in the prepared hybrid electrode.

**Fig. S5** TEM images of the resulting hybrid electrode with an increasing Ni/Ru ratio (scale = 10 nm).
**Fig. S6** XPS Survey Scans of the pristine Ru reference, the prepared hybrid electrodes (Ni/Ru ratios of 0.36 and 1.63), and an individually prepared Ni₃P.

**Fig. S7** Ru K-edge XANES spectra for the hybrid electrode (Ni/Ru ratio of 0.36) and comparison with a Ru foil reference.
Fig. 58 Required overpotentials during OER assessment of the pristine Ru reference in either alkaline 0.1 M KOH or acidic 0.5 M H$_2$SO$_4$ electrolytes at 25 °C, at a scan rate of 5 mV/s.