

## Electronic Supplementary Information

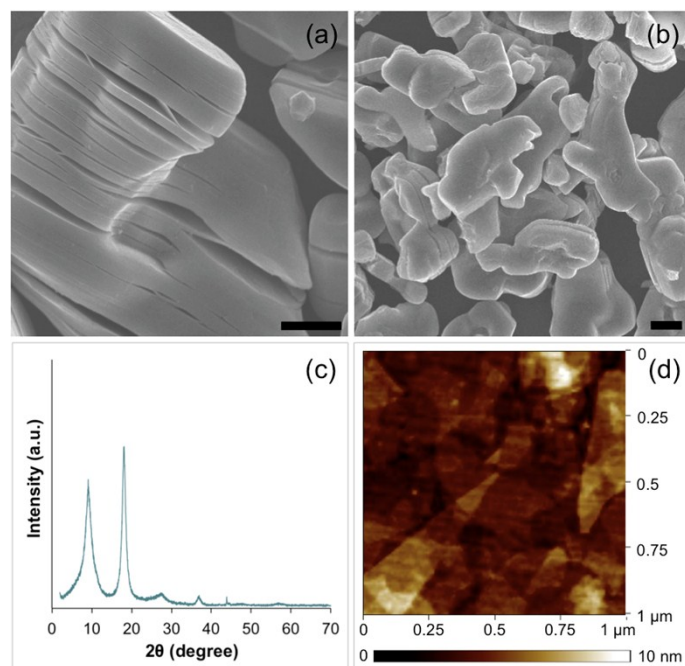
### **Arising Synergetic and Antagonistic Effects in the Design of Ni- and Ru-Based Water Splitting Electrocatalysts**

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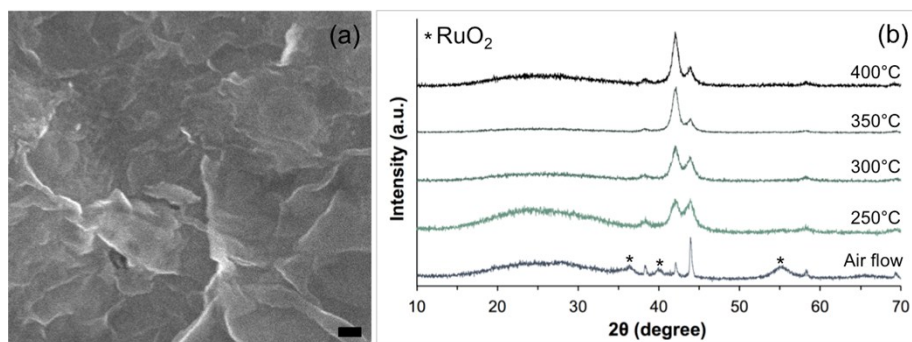
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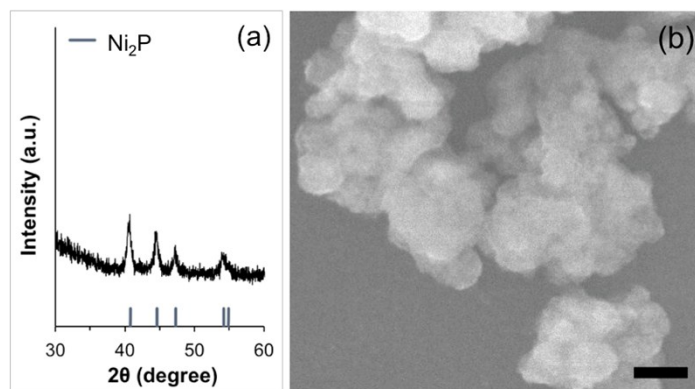
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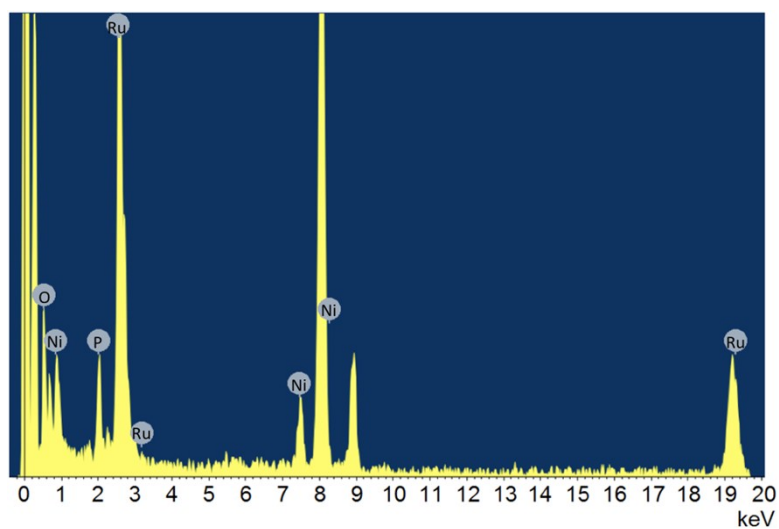
**Fig. S1** SEM images of (a) the pristine sodium ruthenate following heating at 900°C (scale = 1 μm) and (b) the hydrated layered protonic ruthenate following proton exchange with HCl 1M (scale = 1 μm). (c) XRD patterns of the final TMA-intercalated exfoliated material. (d) AFM analysis of the resulting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



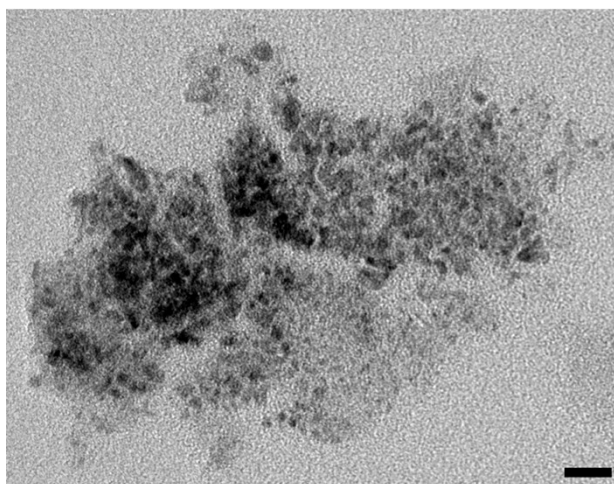
**Fig. S2** (a) SEM images (scale = 100 nm) of pristine Ru-based electrode following heat treatment at 250°C under argon. (b) XRD patterns for pristine Ru-based references heat treated under argon flow at an increasing 250°C to 400°C temperature range, and alternatively following treatment under air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



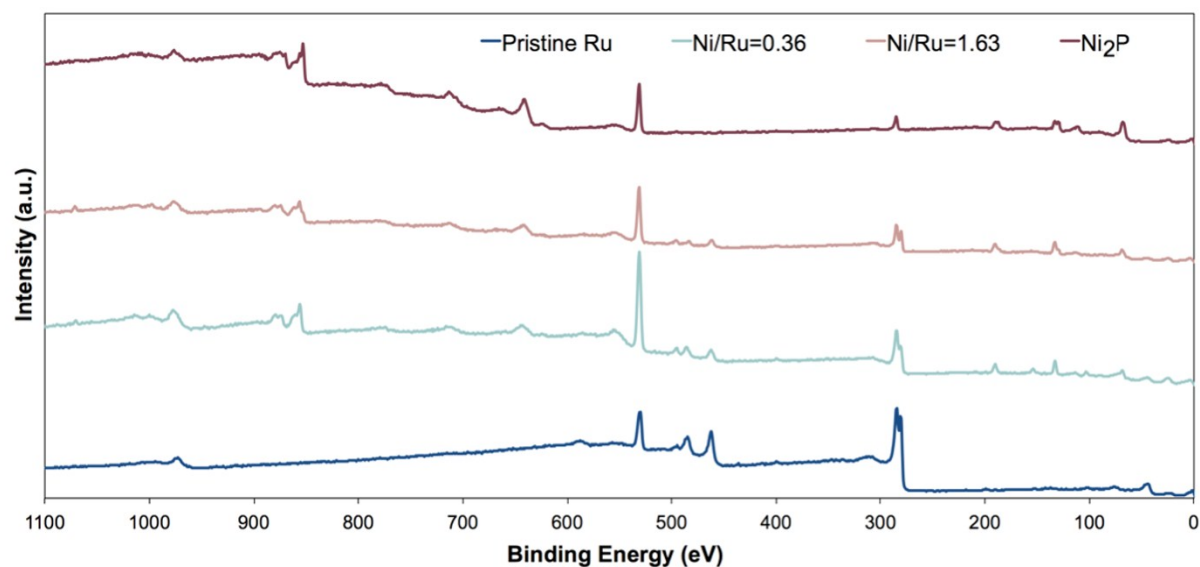
**Fig. S3** (a) XRD patterns of individually synthesized  $\text{Ni}_2\text{P}$  at  $250^\circ\text{C}$  under Ar flow and (b) corresponding SEM images (scale = 100 nm).



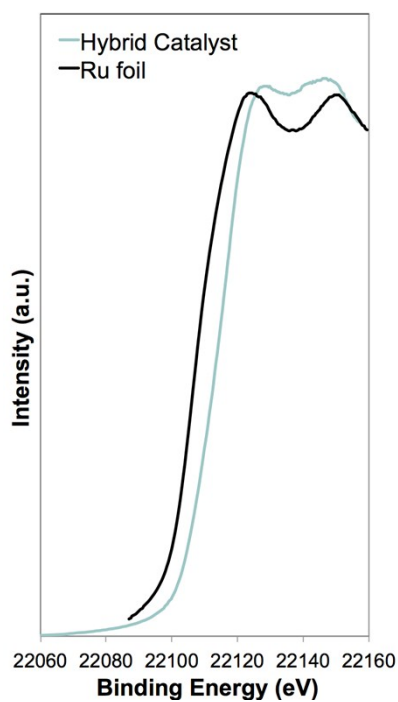
**Fig. S4** EDX results evidencing the presence of Ru, Ni, and P in the prepared hybrid electrode.



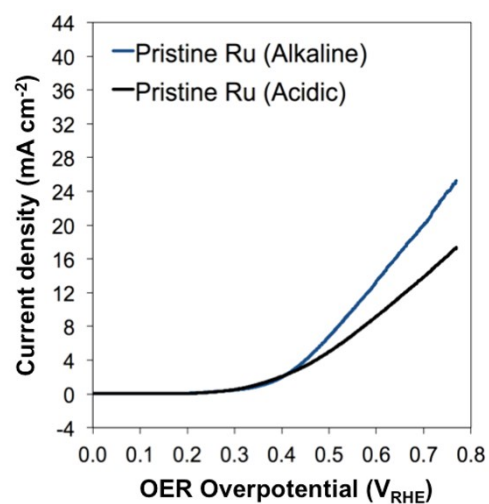
**Fig. S5** TEM images of the resulting hybrid electrode with an increasing Ni/Ru ratio (scale = 10 nm).



**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 Ni<sub>2</sub>P.



**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. S8** Required overpotentials during OER assessment of the pristine Ru reference in either alkaline 0.1 M KOH or acidic 0.5 M H<sub>2</sub>SO<sub>4</sub> electrolytes at 25 °C, at a scan rate of 5 mV/s.