

## Electronic Supplementary Material

### **Atomically ultrathin RhCo alloy nanosheets aggregates for efficient water electrolysis in broad pH range**

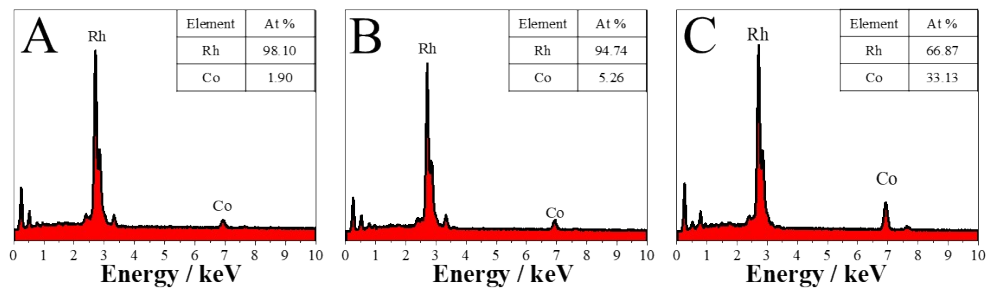
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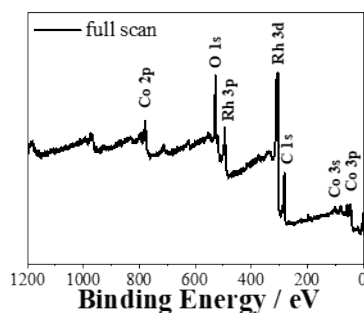
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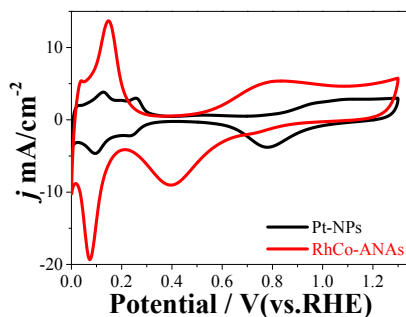
E-mail: ndchenyu@gmail.com (Y. Chen).



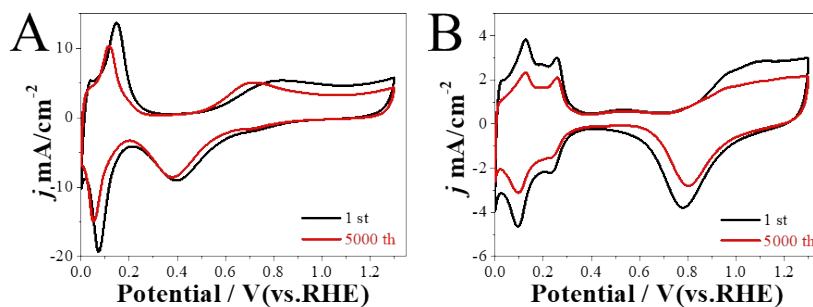
**Fig. S1** EDX spectra of RhCo-ANAs obtained at (A) 160 °C, (B) 200 °C, and 240 °C, respectively.



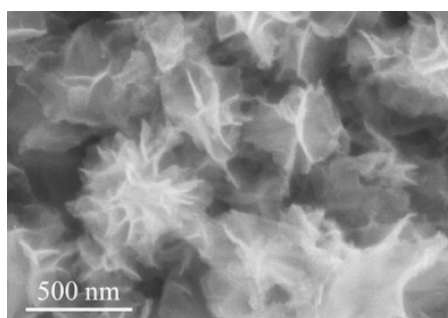
**Fig. S2** XPS survey spectrum of RhCo-ANAs.



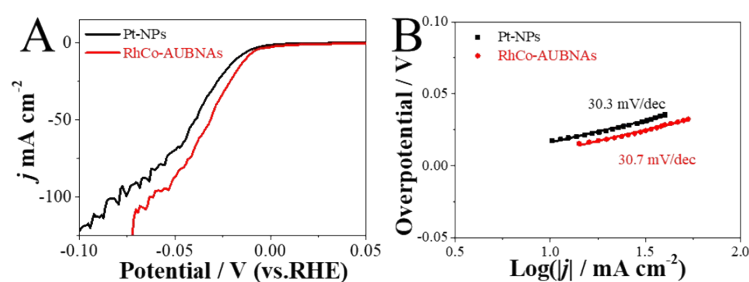
**Fig. S3** CV curves of RhCo-ANAs and Pt-NPs in Ar-saturated 0.5 M H<sub>2</sub>SO<sub>4</sub> solution at 50 mV s<sup>-1</sup>.



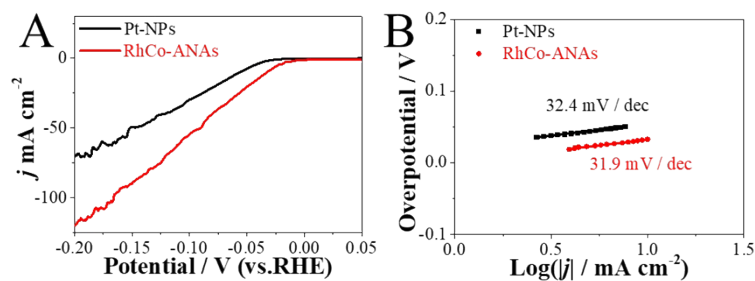
**Fig. S4** CV curves of (A) RhCo-ANAs and (B) Pt-NPs in Ar-saturated 0.5 M H<sub>2</sub>SO<sub>4</sub> solution at 50 mV s<sup>-1</sup> before and after 5 000 scan cycles.



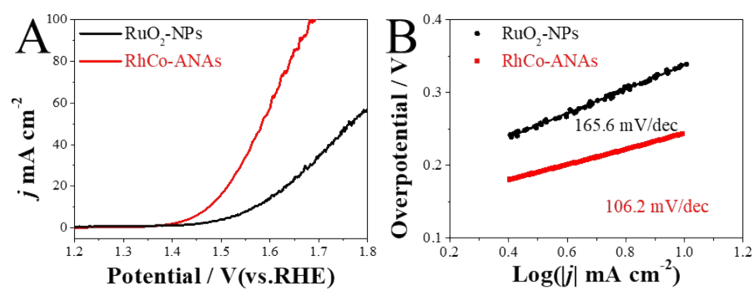
**Fig. S5** SEM image of RhCo-ANAs after 5 000 scan cycles.



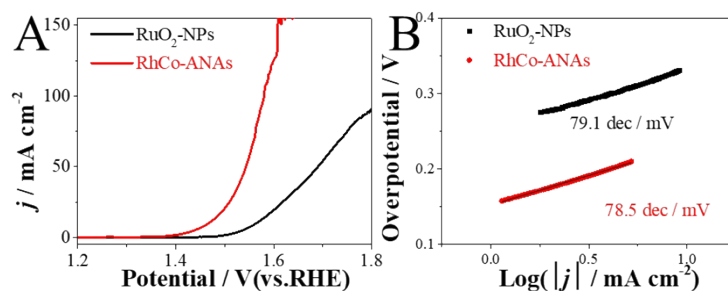
**Fig. S6** (A) HER polarization curves of RhCo-ANAs and Pt-NPs in Ar-saturated 0.5 M  $\text{H}_2\text{SO}_4$  solution at  $5 \text{ mV s}^{-1}$ . (B) Tafel plots of the HER at RhCo-ANAs and Pt-NPs in Ar-saturated 0.5 M  $\text{H}_2\text{SO}_4$  solution.



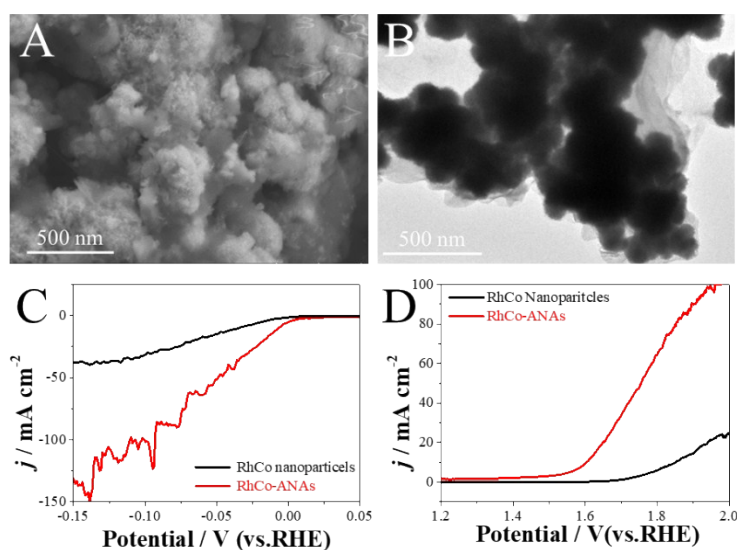
**Fig. S7** (A) HER polarization curves of RhCo-ANAs and Pt-NPs in Ar-saturated 1.0 M KOH solution at  $5 \text{ mV s}^{-1}$ . (B) Tafel plots of HER at RhCo-ANAs and Pt-NPs in Ar-saturated 1.0 M KOH solution.



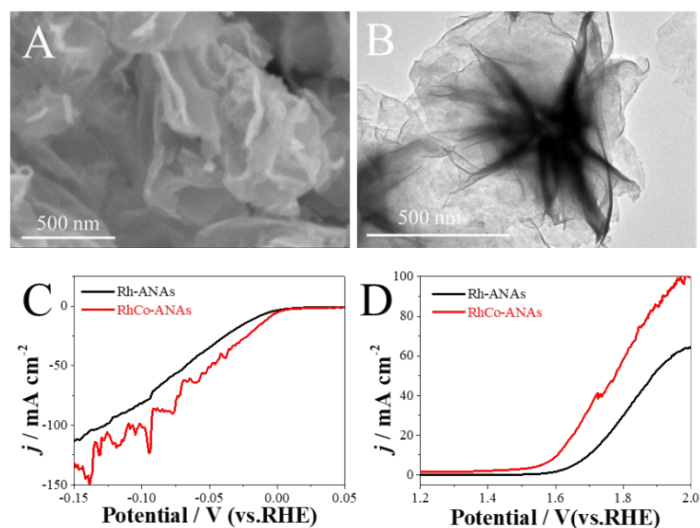
**Fig. S8** (A) OER polarization curves of RhCo-ANAs and  $\text{RuO}_2$ -NPs in Ar-saturated 0.5 M  $\text{H}_2\text{SO}_4$  solution at  $5 \text{ mV s}^{-1}$  and (B) Tafel plots of RhCo-ANAs and  $\text{RuO}_2$ -NPs.



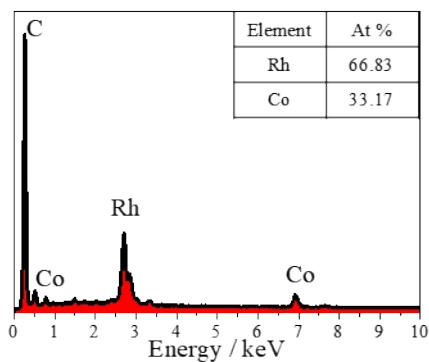
**Fig. S9** (A) OER polarization curves of RhCo-ANAs and RuO<sub>2</sub>-NPs in Ar-saturated 1.0 M KOH solution at 5 mV s<sup>-1</sup> and (B) Tafel plots of RhCo-ANAs and RuO<sub>2</sub>-NPs.



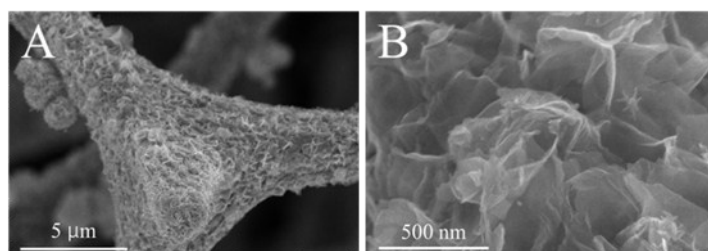
**Fig. S10.** (A) SEM and (B) TEM images of spherical RhCo nanoparticles. (C) HER polarization curves of RhCo-ANAs and spherical RhCo nanoparticles in Ar-saturated 1.0 M PBS solution at 5 mV s<sup>-1</sup>. (D) OER polarization curves of RhCo-ANAs and spherical RhCo nanoparticles in Ar-saturated 1.0 M PBS solution at 5 mV s<sup>-1</sup>. Herein, spherical RhCo nanoparticles are obtained by using RhCl<sub>3</sub> and CoCl<sub>2</sub> as reaction precursors and HCHO as reducing agent at 240 °C for 6 h.



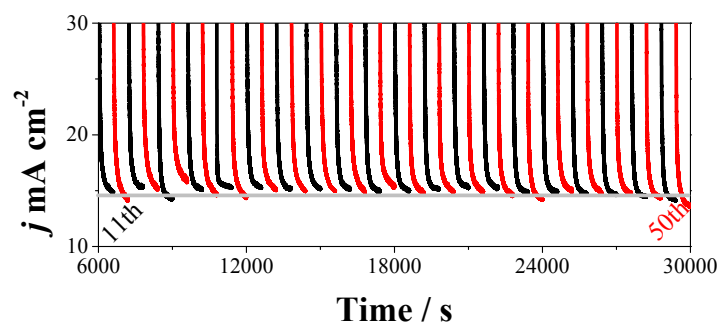
**Fig. S11** (A) SEM and (B) TEM images of Rh-ANAs. (C) HER polarization curves of RhCo-ANAs and monometallic Rh-ANAs in Ar-saturated 1.0 M PBS solution at 5 mV s<sup>-1</sup>. (D) OER polarization curves of RhCo-ANAs and monometallic Rh-ANAs in Ar-saturated 1.0 M PBS solution at 5 mV s<sup>-1</sup>. Herein, Rh-ANAs was obtained by selecting reaction temperature at 160 °C (Fig. S1A)



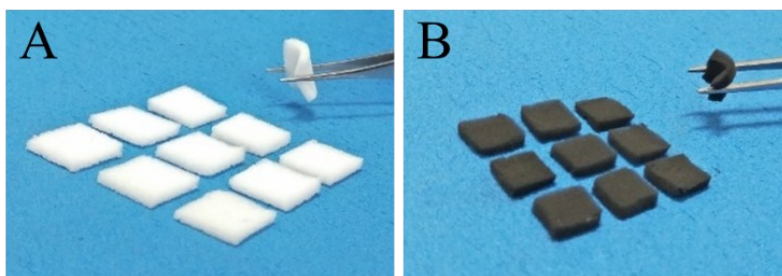
**Fig. S12** EDX spectrum of RhCo-ANAs/CF.



**Fig. S13** SEM images of RhCo-ANAs/CF after the stability test (A) SEM images of RhCo-ANAs/CF, (B) magnified SEM images of RhCo-ANAs/CF.



**Fig. S14** Chronoamperometric curves of symmetric RhCo-ANAs/CF||RhCo-ANAs/CF electrolyzer in 1.0 M PBS solution in periodically switching cathode and anode mode. Applied voltage: 1.65 V; Switching time interval: 10 min.



**Fig. S15** The photograph of (A) sponge scouring pad and (B) CF with flexible property.