# Wire-on-flake Heterostructured Ternary $Co_{0.5}Ni_{0.5}P$/CC: An Efficient Hydrogen Evolution Electrocatalyst

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**KEYWORDS:** Wire-on-flake Heterostructure, Ternary $Co_{0.5}Ni_{0.5}P$/CC, Self-supported electrode, Hydrogen evolution

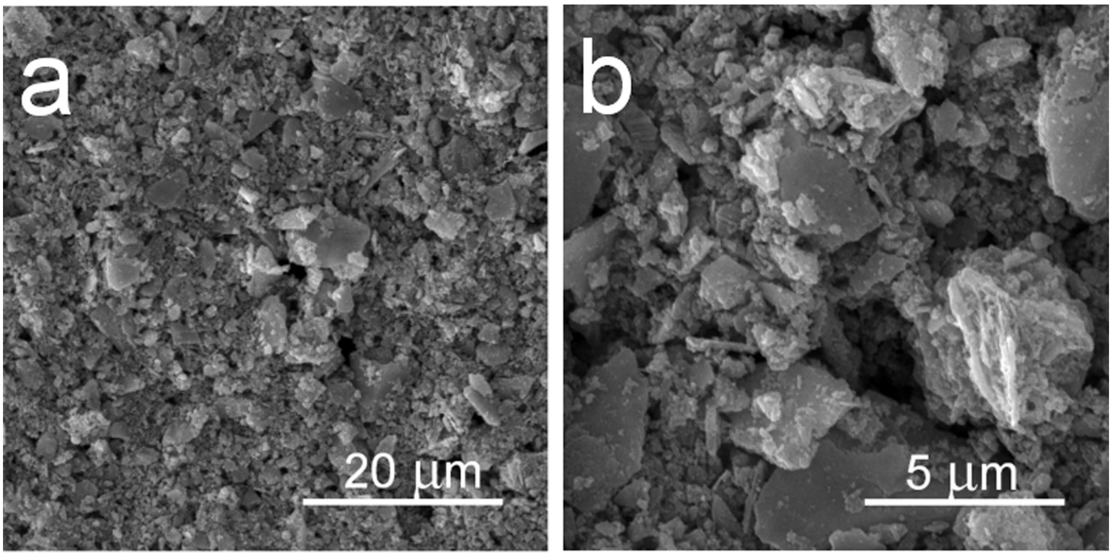


**Figure S1.** SEM images of $Co_{0.5}Ni_{0.5}P$ scratched from CC with different magnifications.

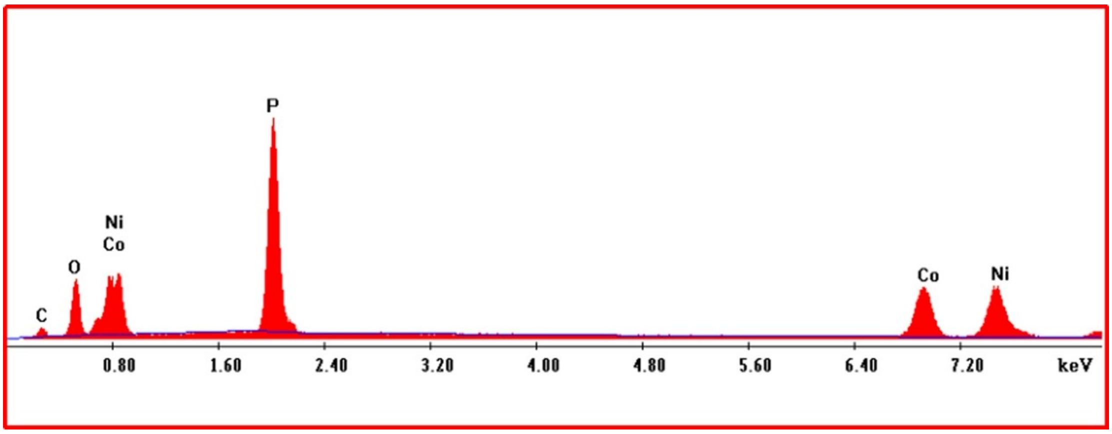


**Figure S2.** EDX spectrum for $Co_{0.5}Ni_{0.5}P$/CC.

**Table S1.** Elements percentage of $Co_{0.5}Ni_{0.5}P$/CC obtained from EDX and ICP-MS.

| Elements | Weight% (EDX) | Atomic% (EDX) | Atomic% (ICP-MS) |
|---|---|---|---|
| Co | 33.13 | 25.48 | 25.91 |
| Ni | 33.73 | 26.04 | 25.35 |
| P | 33.14 | 48.48 | 48.74 |
| Total | 100 | 100 | 100 |
| Co:Ni:P | | 1:1.02:1.90 | 1.02:1:1.92 |

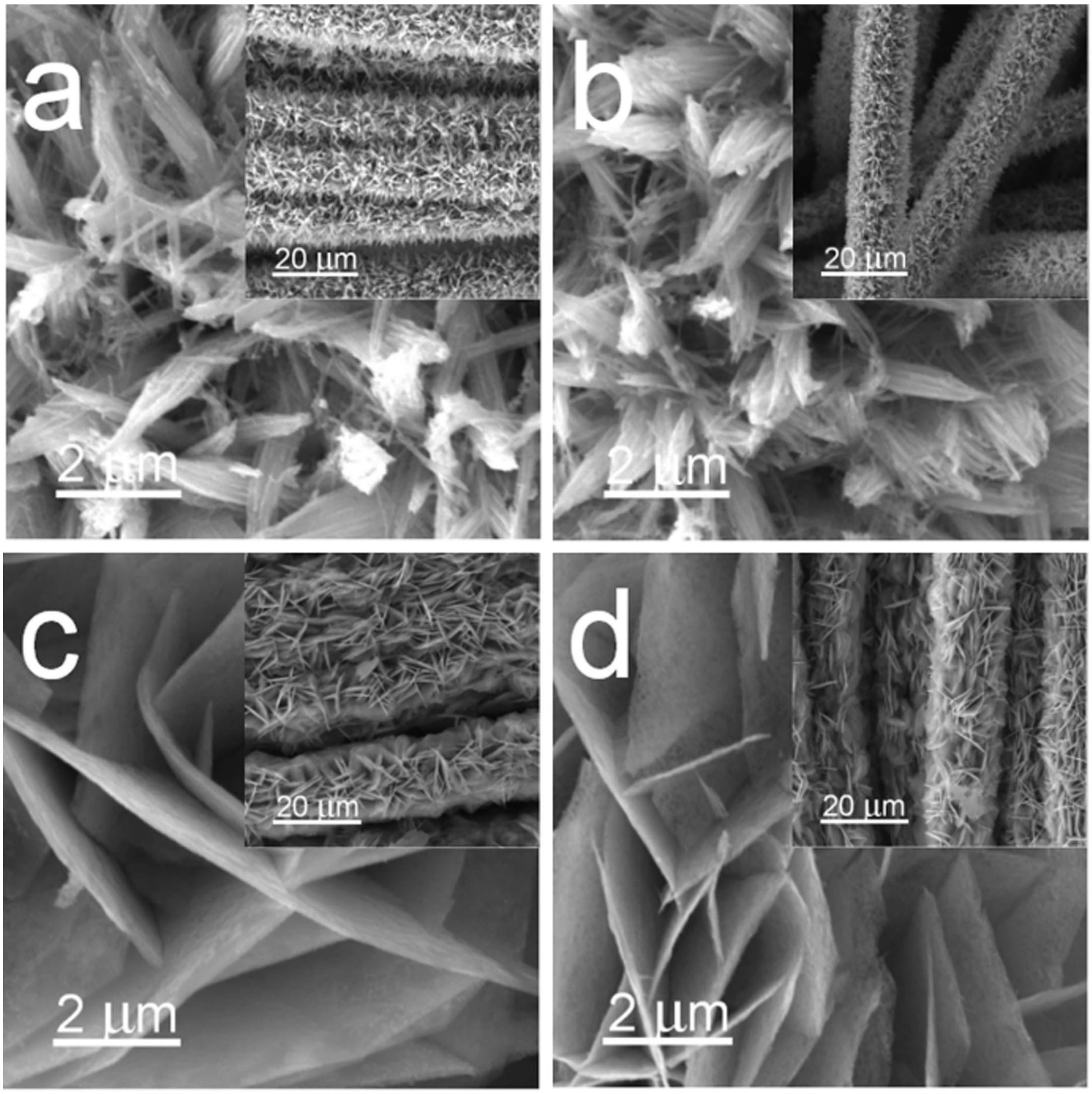


**Figure S3.** SEM images for (a) $Co_{0.75}Ni_{0.25}P$/CC, (b) $Co_{0.66}Ni_{0.33}P$/CC, (c) $Co_{0.33}Ni_{0.66}P$/CC and (d) $Co_{0.25}Ni_{0.75}P$/CC. Inset in (a-d): SEM images of $Co_{0.75}Ni_{0.25}P$/CC, $Co_{0.66}Ni_{0.33}P$/CC, $Co_{0.33}Ni_{0.66}P$/CC and $Co_{0.25}Ni_{0.75}P$/CC with a larger magnification, respectively.

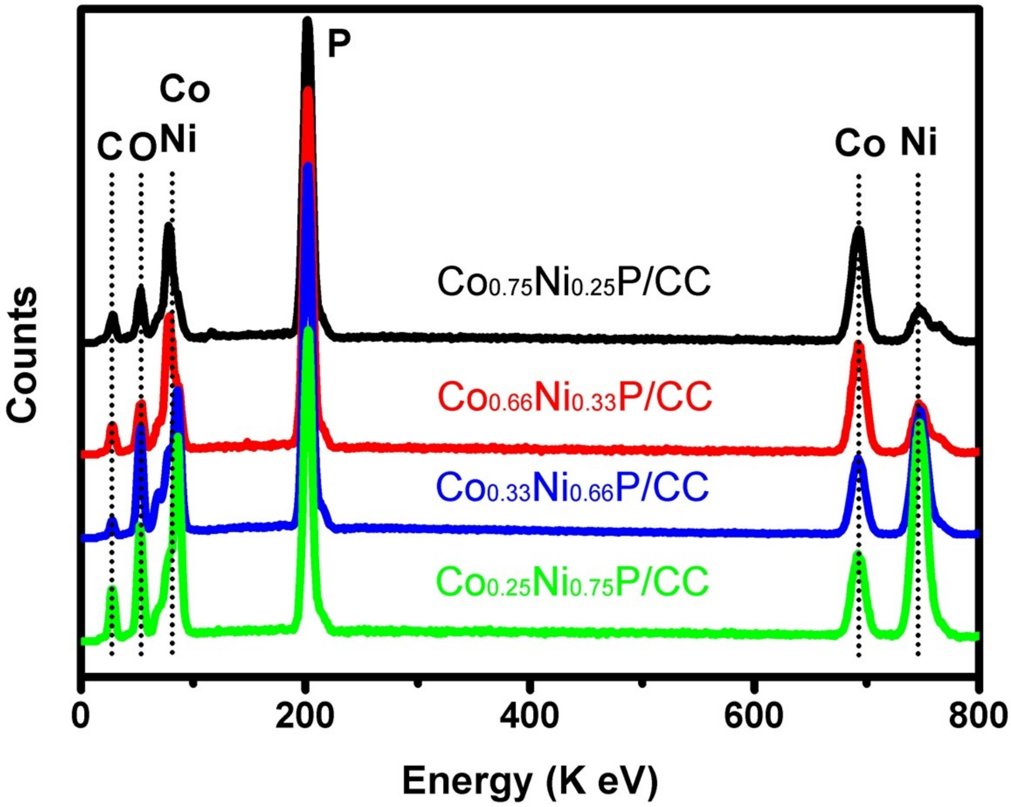


**Figure S4.** EDX spectra for $Co_{0.75}Ni_{0.25}P$/CC, $Co_{0.66}Ni_{0.33}P$/CC, $Co_{0.33}Ni_{0.66}P$/CC and $Co_{0.25}Ni_{0.75}P$/CC, respectively.

**Table S2.** Elements percentage of $Co_{0.75}Ni_{0.25}P$/CC, $Co_{0.66}Ni_{0.33}P$/CC, $Co_{0.33}Ni_{0.66}P$/CC and $Co_{0.25}Ni_{0.75}P$/CC obtained from EDX.

| **Elements** | **Atomic% (EDX)** | | | |
|---|---|---|---|---|
| | $Co_{0.75}Ni_{0.25}P$/CC | $Co_{0.66}Ni_{0.33}P$/CC | $Co_{0.33}Ni_{0.66}P$/CC | $Co_{0.25}Ni_{0.75}P$/CC |
| Co | 50.33 | 50.7 | 6.27 | 3.48 |
| Ni | 36.86 | 32.57 | 21.14 | 14.71 |
| P | 12.81 | 16.73 | 72.59 | 81.81 |
| Total | 100 | 100 | 100 | 100 |
| Co:Ni:P | 2.88:1:4 | 1.95:1:3 | 3.4:11.6:1 | 4.23:23.5:1 |

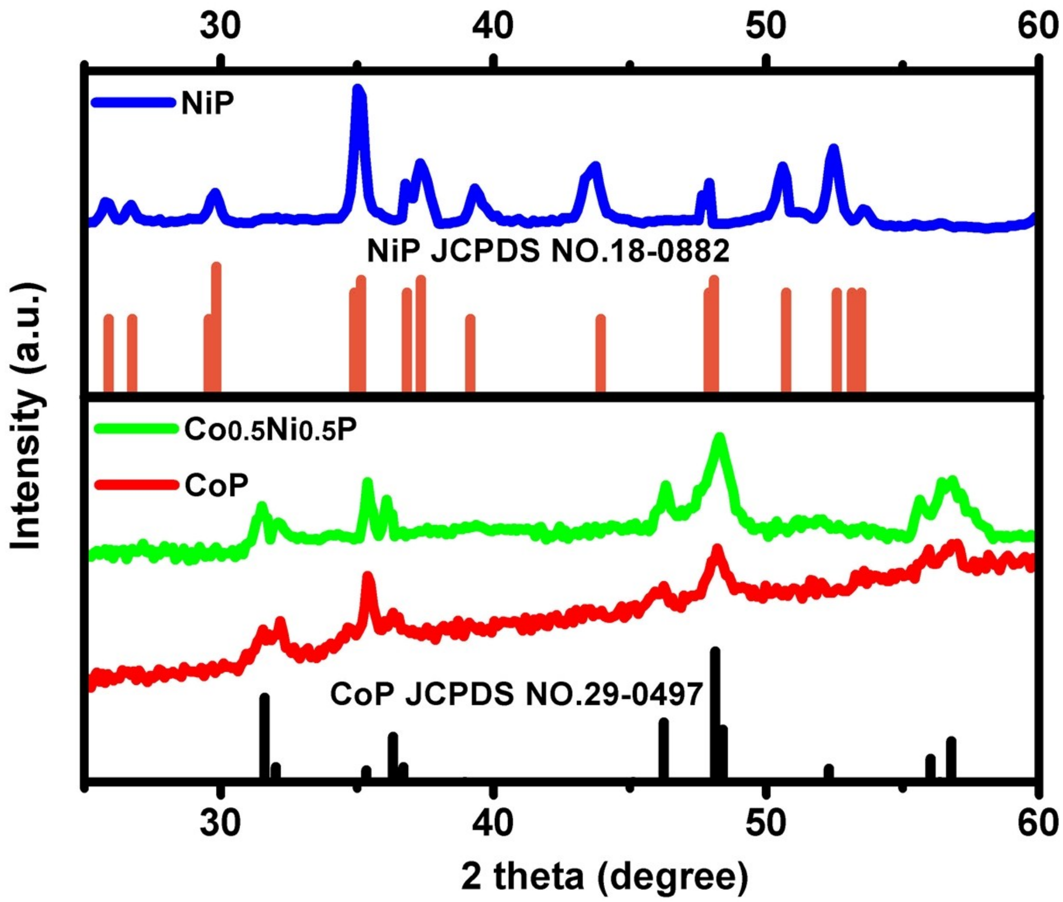


**Figure S5.** XRD spectra for NiP, CoP and $Co_{0.5}Ni_{0.5}P$ scratched from CC.

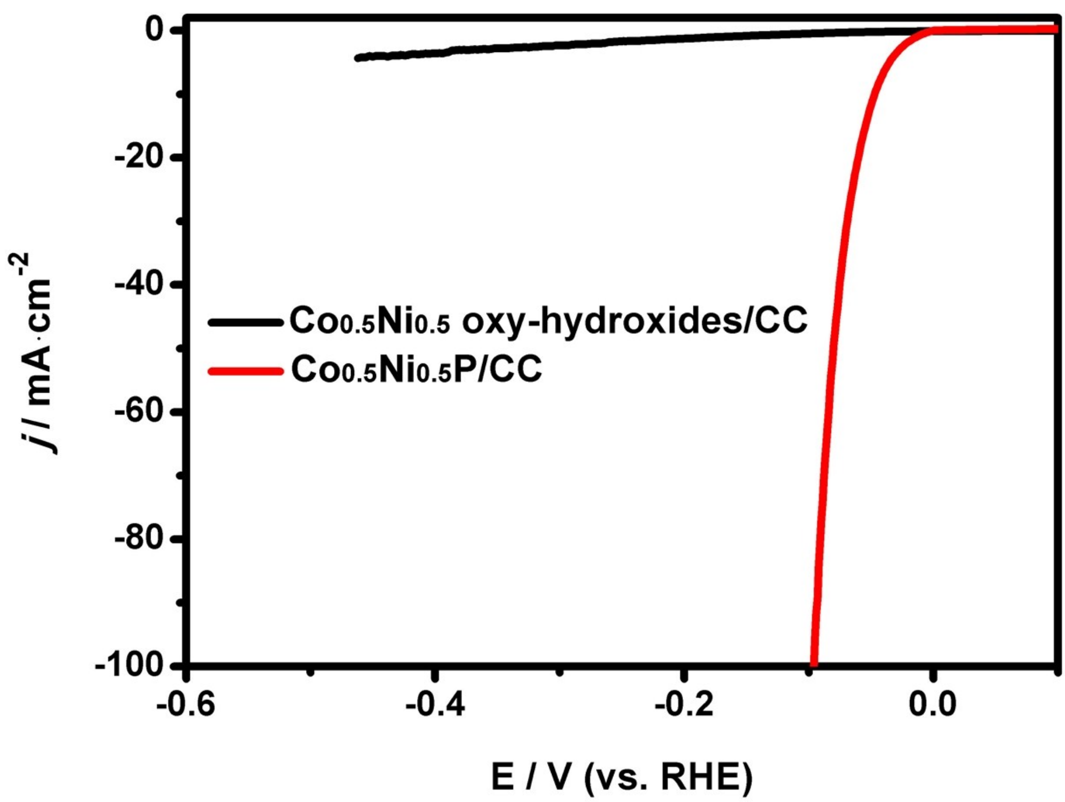


**Figure S6.** LSV curves for $Co_{0.5}Ni_{0.5}$ oxy-hydroxides/CC and $Co_{0.5}Ni_{0.5}P$/CC.

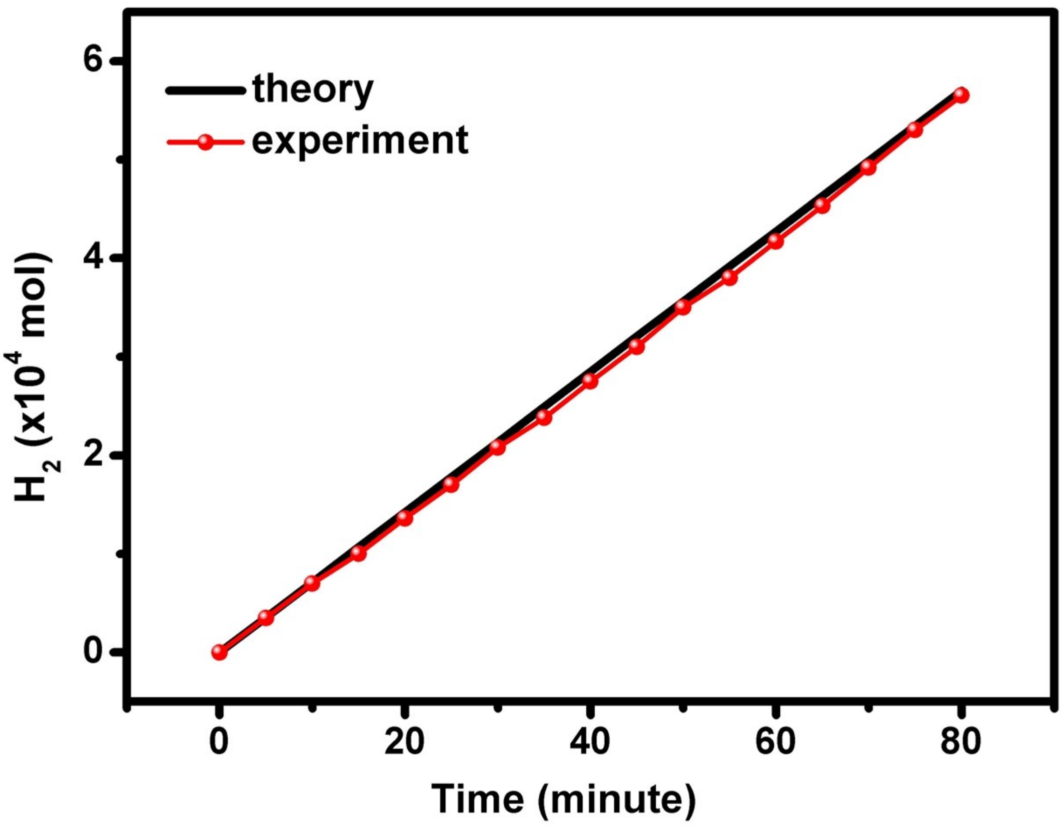


**Figure S7.** The amount of $H_2$ calculated by theory and measured from experiment versus time for $Co_{0.5}Ni_{0.5}P$/CC in 0.5 M $H_2SO_4$.

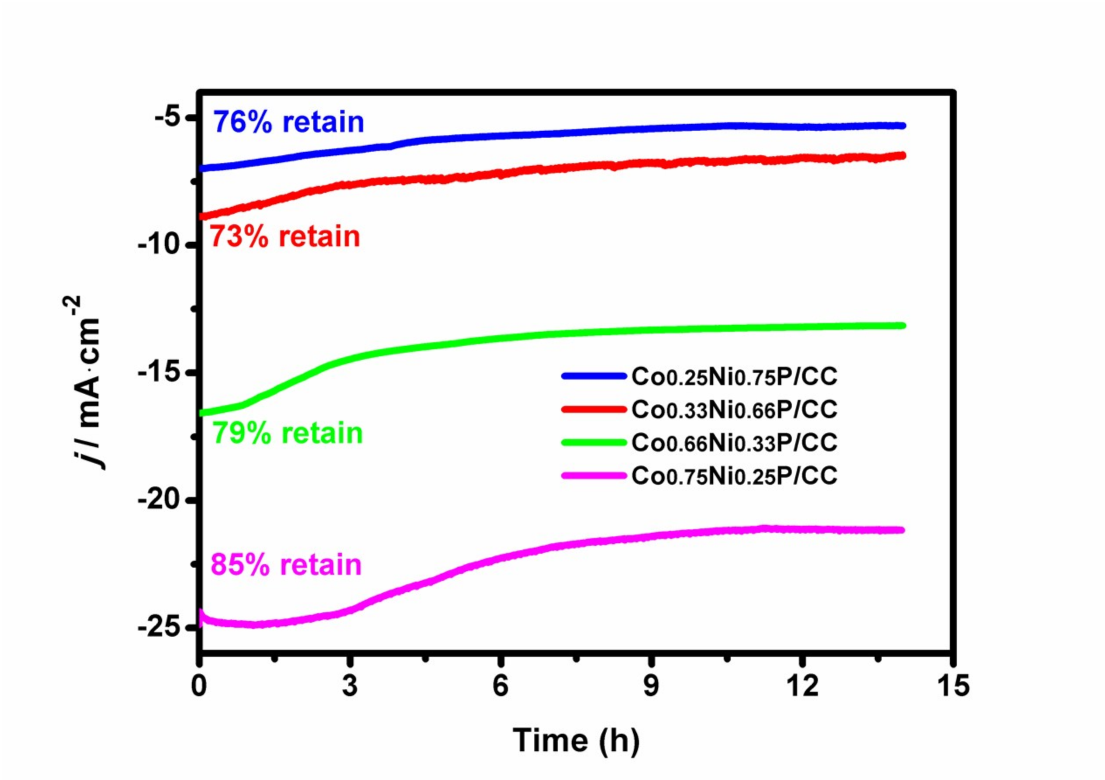


**Figure S8.** Time-dependent current density curves of $Co_{0.25}Ni_{0.75}P$/CC, $Co_{0.33}Ni_{0.66}P$/CC, $Co_{0.66}Ni_{0.33}P$/CC and $Co_{0.75}Ni_{0.25}P$/CC under a fixed overpotential of 90 mV for 14 h.

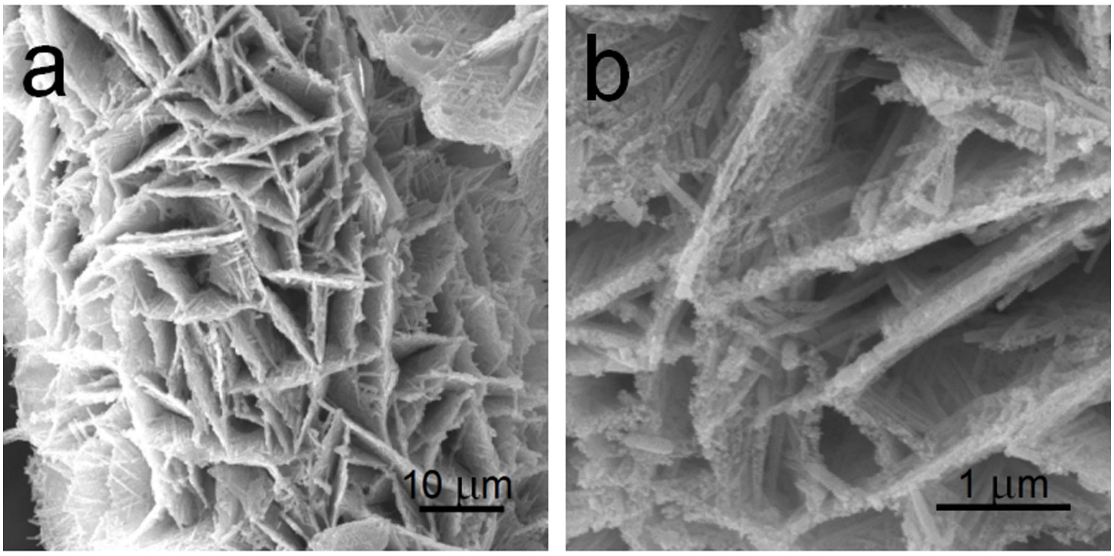


**Figure S9.** (a) and (b) SEM images of $Co_{0.5}Ni_{0.5}P$/CC after time-dependent current density test under different magnifications.

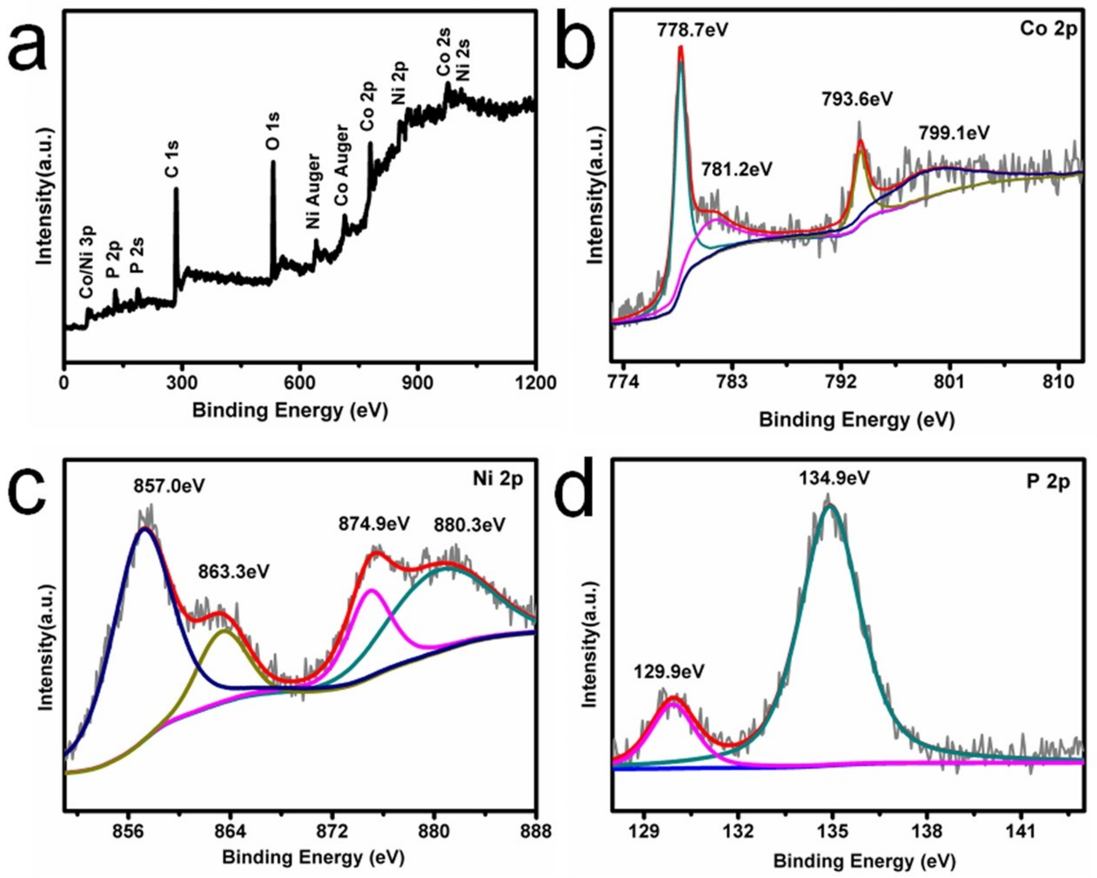


**Figure S10.** (a) XPS characterization of $Co_{0.5}Ni_{0.5}P$/CC and its corresponding (b) Co 2p, (c) Ni 2p and (d) P 2p spectra after time-dependent current density test.

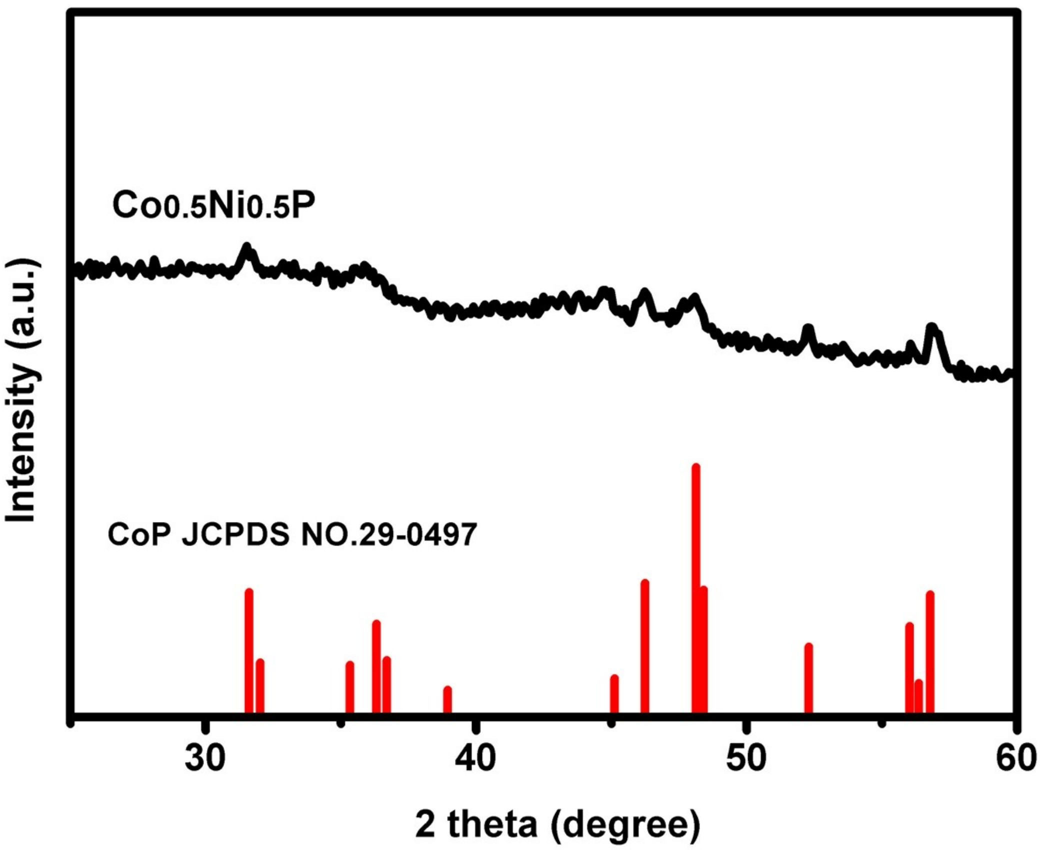


**Figure S11.** XRD spectrum of $Co_{0.5}Ni_{0.5}P$ scratched from CC after time-dependent current density test.

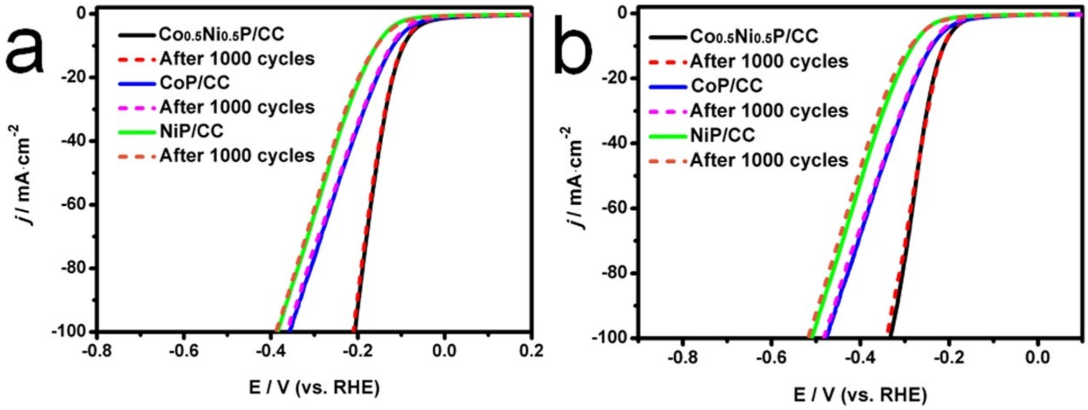


**Figure S12.** (a) Polarization curves of $Co_{0.5}Ni_{0.5}P$/CC, CoP/CC and NiP/CC before and after 1000 CV cycles in (a) 0.5 M PBS and (b) 1.0 M KOH.

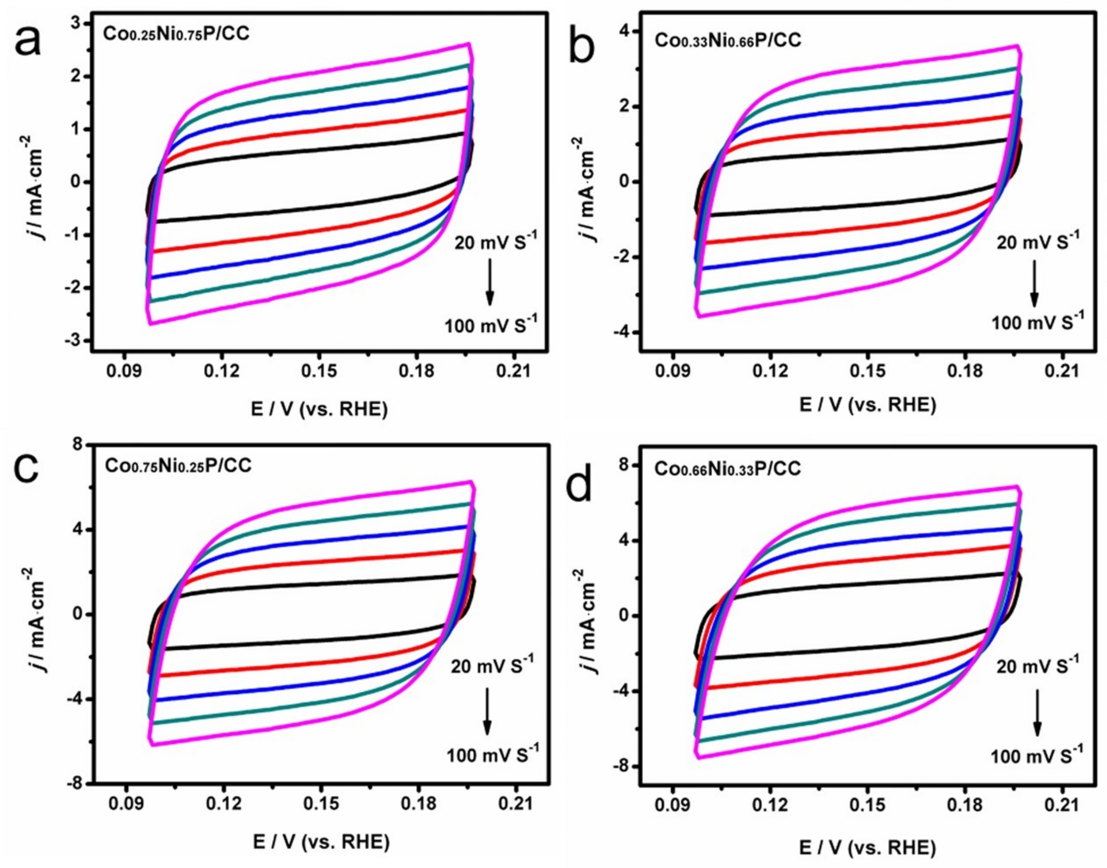


**Figure S13.** Cyclic voltammetry curves for (a) $Co_{0.25}Ni_{0.75}P$/CC, (b) $Co_{0.33}Ni_{0.66}P$/CC, (c) $Co_{0.75}Ni_{0.25}P$/CC and (d) $Co_{0.66}Ni_{0.33}P$/CC at different scan rates.

**Table S3.** Comparison of HER activity in acidic media for $Co_{0.5}Ni_{0.5}P$/CC with other existed non-noble-metal electrocatalysts.

| Catalyst | Tafel slope (mV dec$^{-1}$) | $C_{dl}$ (mF cm$^{-2}$) | Current density (mA cm$^{-2}$) | Corresponding Overpotential (mV) | Ref |
|---|---|---|---|---|---|
| $Co_{0.5}Ni_{0.5}P$/CC | 34.1 | 74.7 | 10 | 47 | This work |
| | | | 100 | 96 | |
| P-1T-$MoS_2$ | 43 | 63.1 | 10 | 153 | S1 |
| $Co_2P$@NPG | 58 | 66.8 | 10 | 103 | S2 |
| | | | 20 | 129 | |
| $Cu_7S_4$@$MoS_2$ | 48 | | 10 | 133 | S3 |
| Co@BCN | 63.7 | 83 | 10 | 96 | S4 |
| Se-enriched $NiSe_2$ | 32 | 10.93 | 10 | 117 | S5 |
| CoPS | 56 | 99.6 | 10 | 48 | S6 |
| Mo2C@NPC/NPRGO | 30 | 17.9 | 10 | 34 | S7 |
| Co-C-N complex | 55 | 400 | 10 | 138 | S8 |
| | | | 100 | 212 | |
| $CoSe_2$ nanoparticle/CP | 40 | 14.1 | 10 | 137 | S9 |
| | | | 100 | 181 | |
| α-INS nanosheets | 40 | | 10 | 105 | S10 |

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