

## Electronic Supporting Information (ESI)

Built-in Electric Field Induced Interfacial Charge Distributions of  
 $\text{Ni}_2\text{P}/\text{NiSe}_2$  Heterojunction for Urea-Assisted Hydrogen Evolution Reaction

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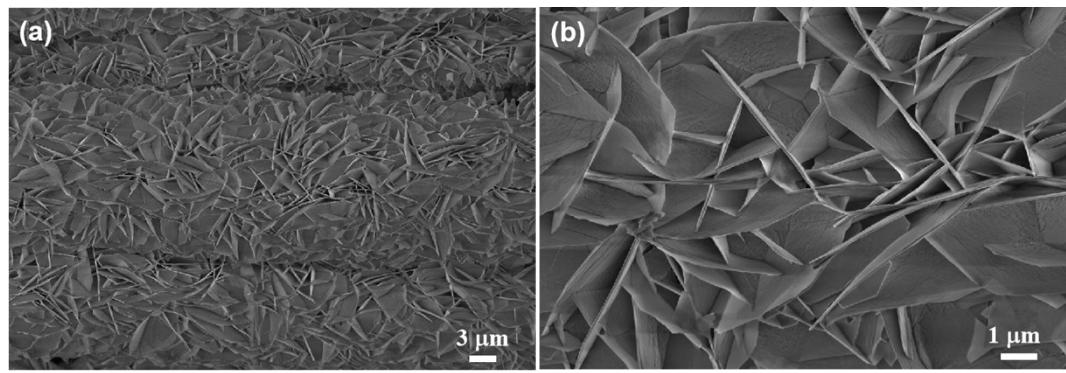
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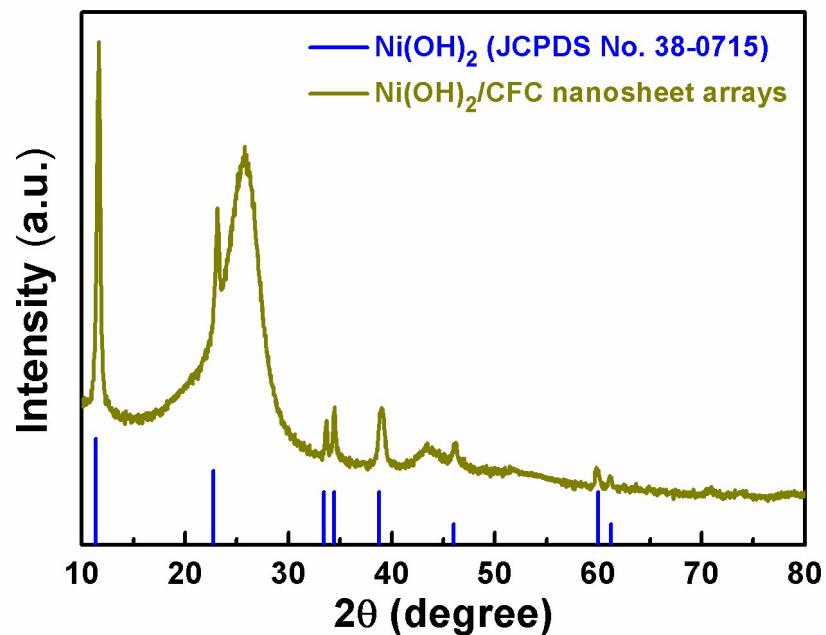
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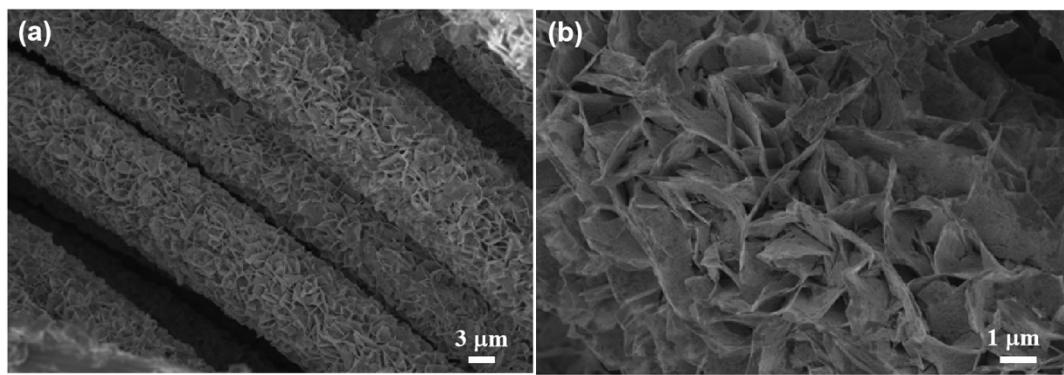
E-mail: ouyangboyi@njust.edu.cn; chm\_shenq@ujn.edu.cn; chm\_licc@ujn.edu.cn



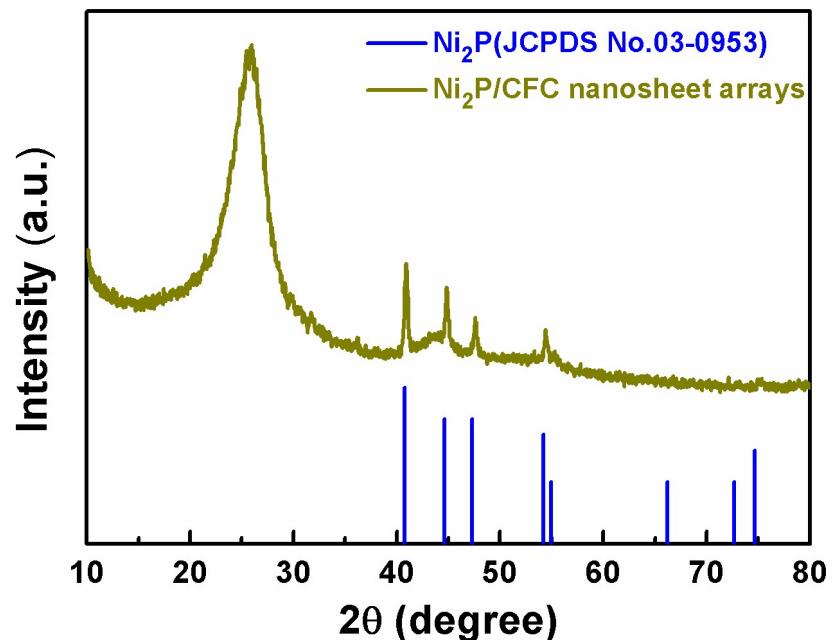
**Fig. S1** (a) Low-magnification SEM image and (b) High-magnification SEM image of  $\text{Ni(OH)}_2/\text{CFC}$  nanosheet arrays precursor.



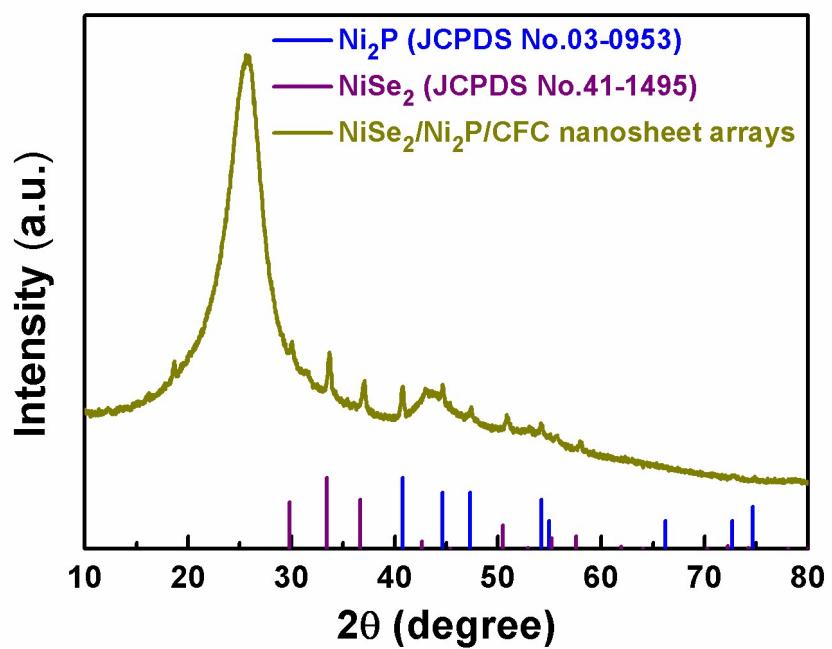
**Fig. S2** XRD pattern of  $\text{Ni(OH)}_2/\text{CFC}$  nanosheet arrays precursor.



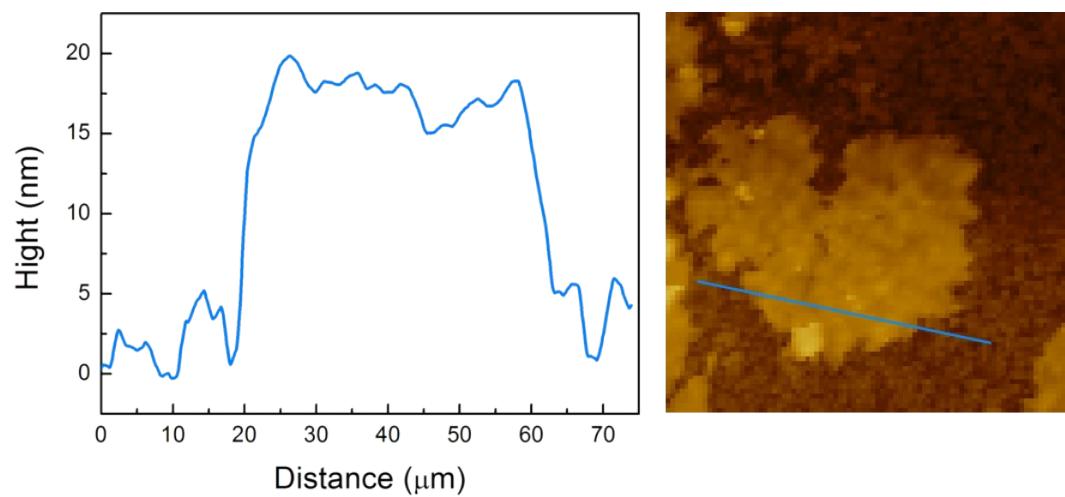
**Fig. S3** (a) Low-magnification SEM image and (b) High-magnification SEM image of  $\text{Ni}_2\text{P}/\text{CFC}$  nanosheet arrays.



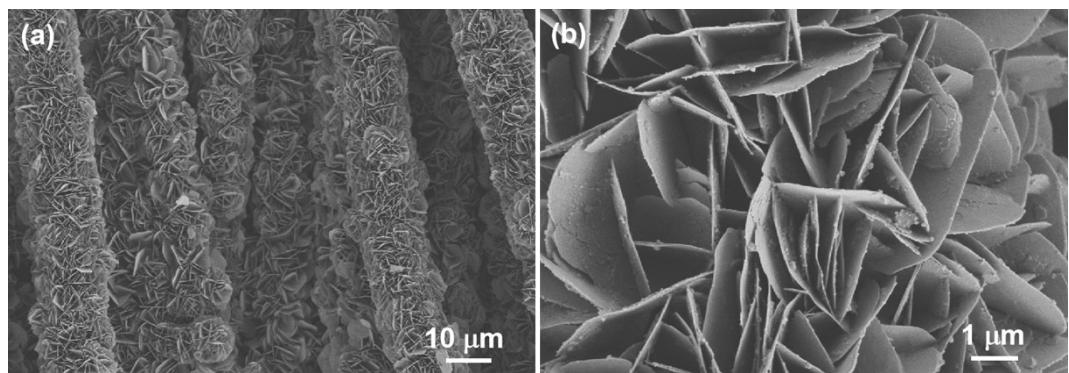
**Fig. S4** XRD pattern of  $\text{Ni}_2\text{P}/\text{CFC}$  nanosheet arrays.



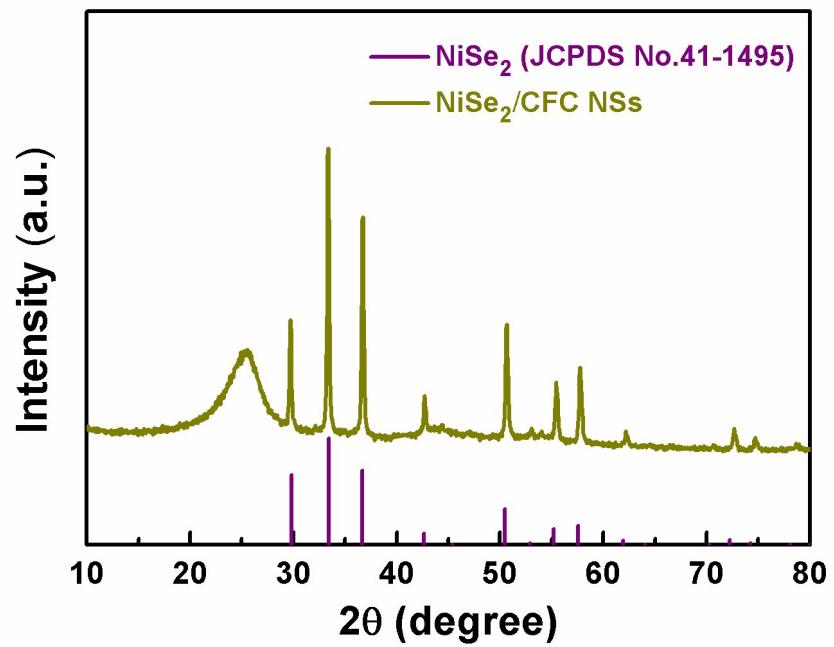
**Fig. S5** XRD pattern of Ni<sub>2</sub>P/NiSe<sub>2</sub>/CFC heterogeneous nanosheet arrays.



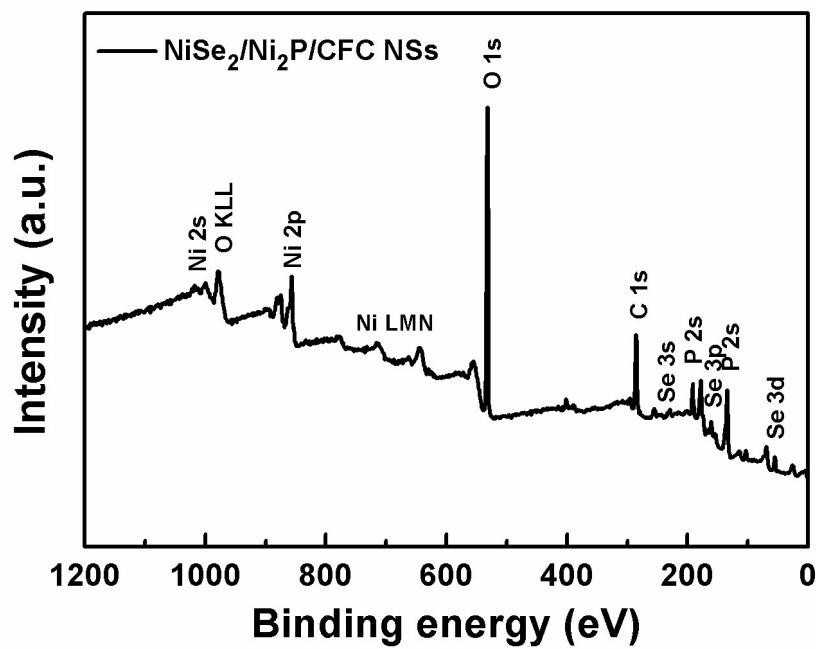
**Fig. S6** AFM image and the corresponding height profiles of Ni<sub>2</sub>P/NiSe<sub>2</sub> nanosheets for the thickness measurements.



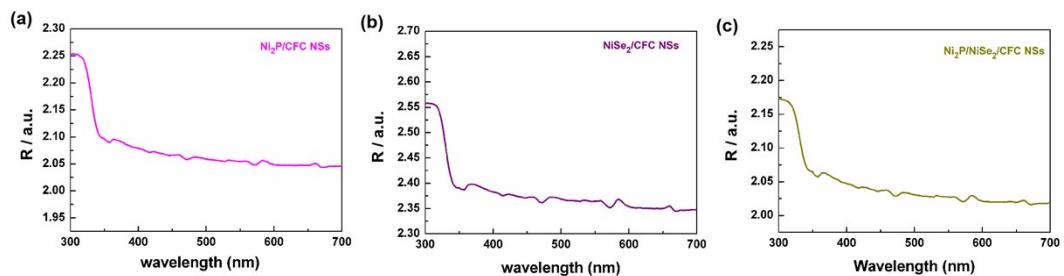
**Fig. S7** (a) Low-magnification SEM image and (b) High-magnification SEM image of  $\text{NiSe}_2/\text{CFC}$  nanosheet arrays.



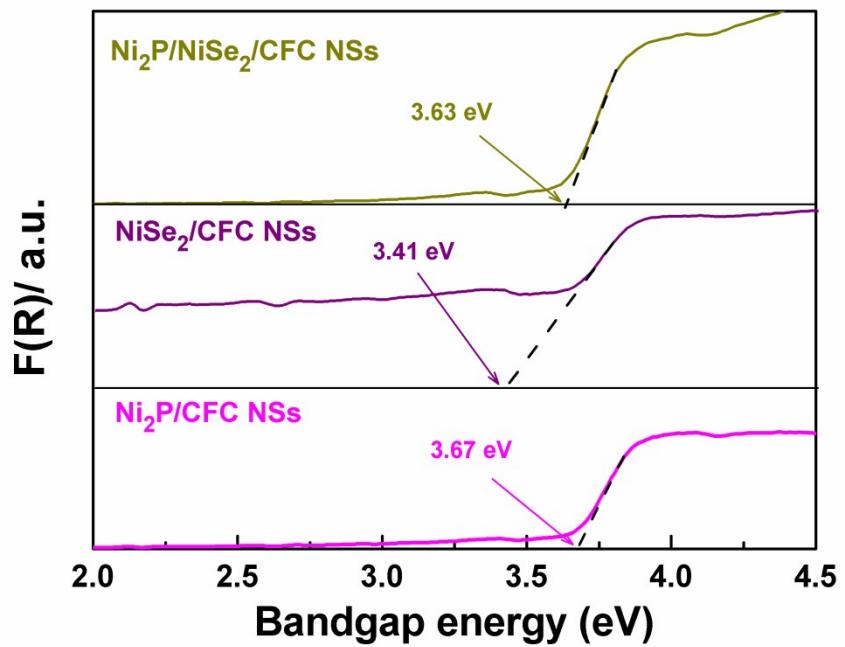
**Fig. S8** XRD pattern of pristine  $\text{NiSe}_2/\text{CFC}$  nanosheet arrays.



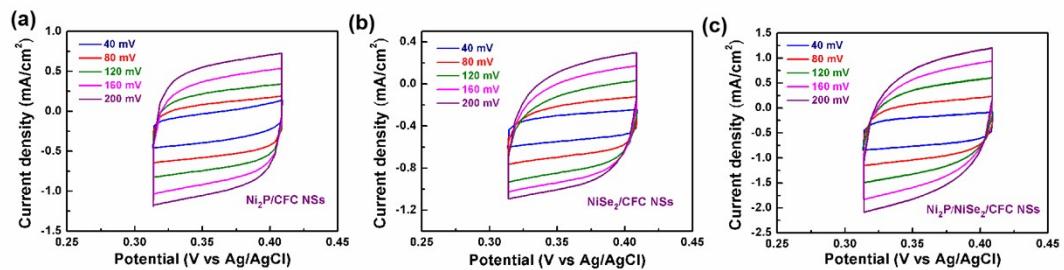
**Fig. S9** The XPS survey spectrum of  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  heterogeneous nanosheet arrays.



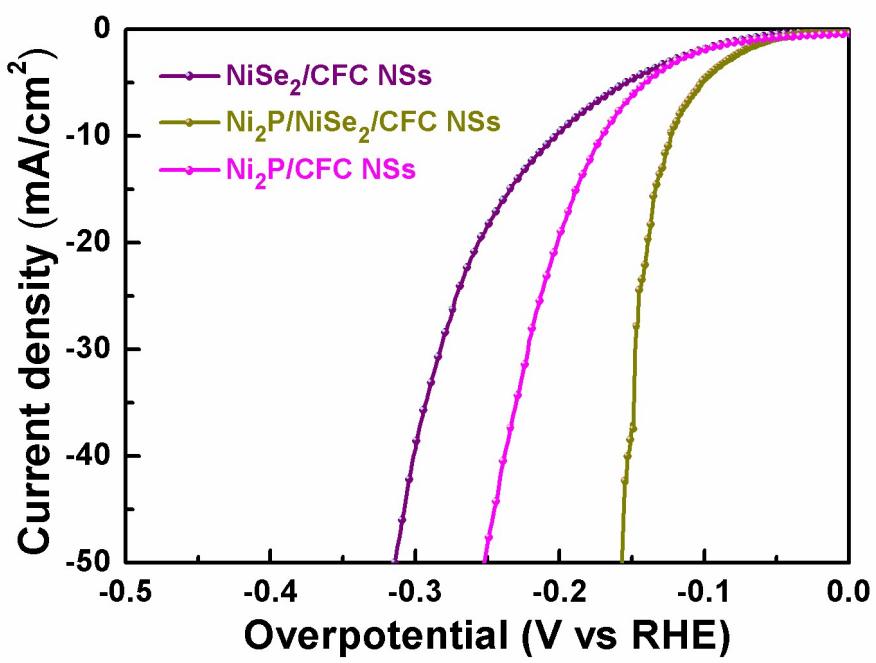
**Fig. S10** UV-VIS diffuse reflectance spectra of (a)  $\text{Ni}_2\text{P}/\text{CFC}$  NSs, (b)  $\text{NiSe}_2/\text{CFC}$  NSs and  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  NSs at different scan rates in 1 M KOH.



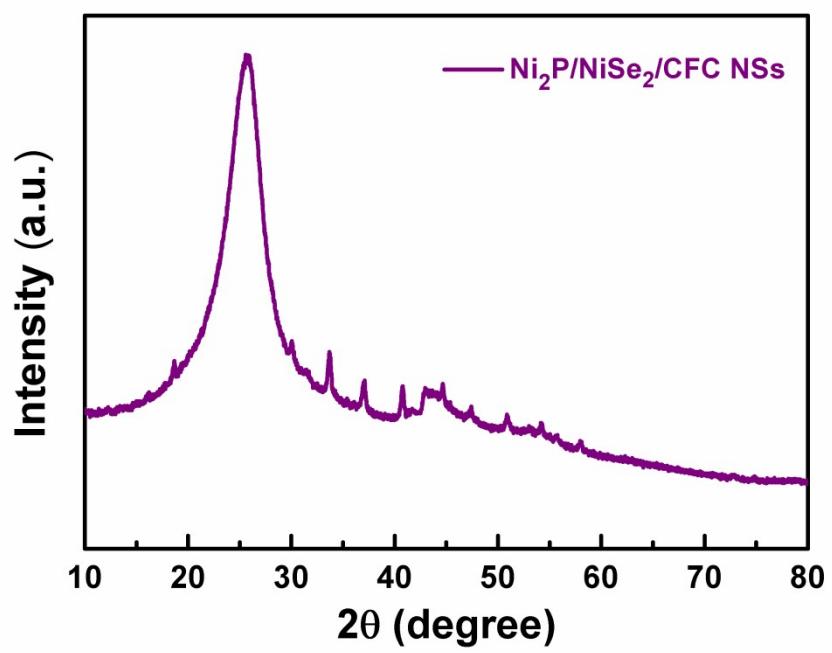
**Fig. S11** UV/Vis spectra.



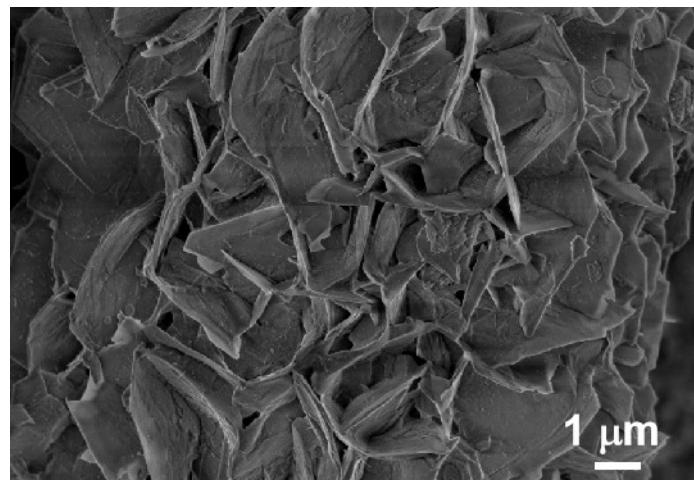
**Fig. S12** Cyclic voltammograms of (a)  $\text{Ni}_2\text{P}/\text{CFC NSs}$ , (b)  $\text{NiSe}_2/\text{CFC NSs}$  and (c)  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC NSs}$  at different scan rates in 1 M KOH.



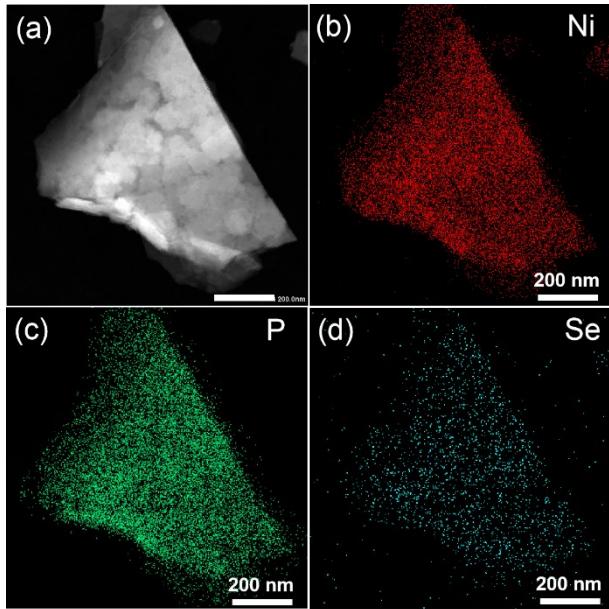
**Fig. S13** HER polarization curves normalized by  $C_{dl}$ .



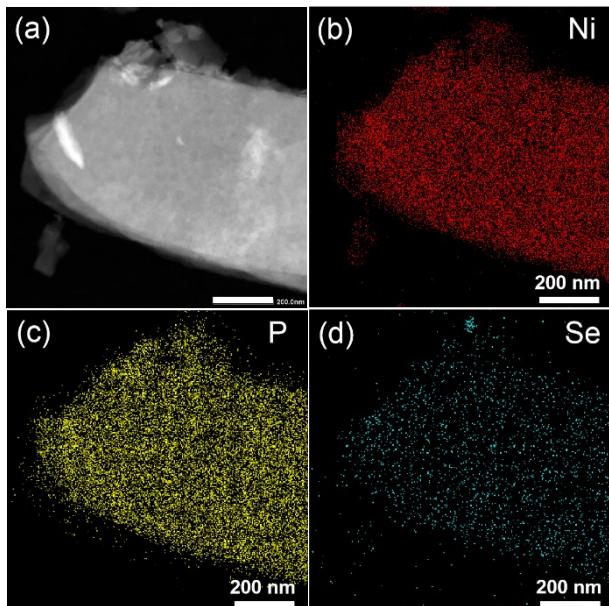
**Fig. S14** XRD pattern of  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  NSs after durability test.



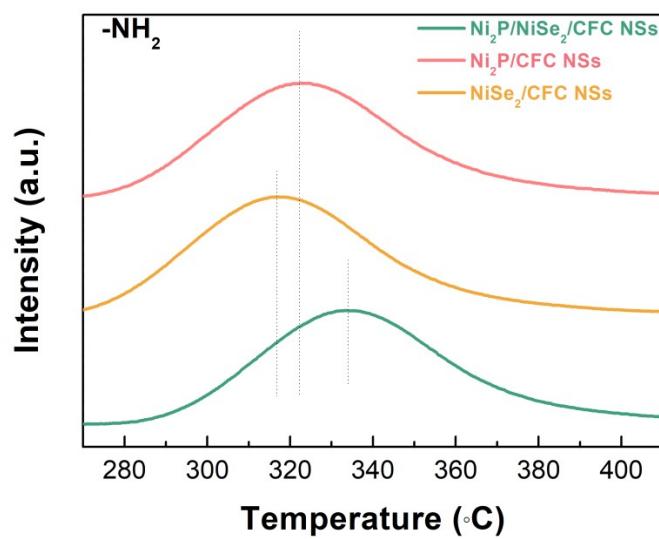
**Fig. S15** SEM image of  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  NSs after durability test.



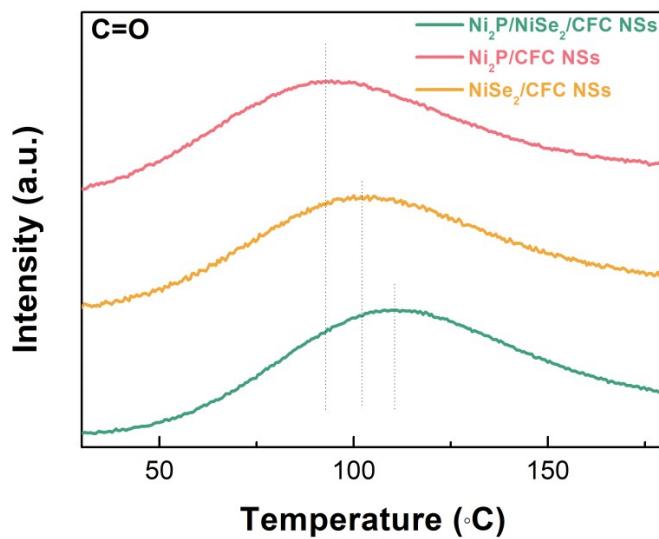
**Fig. S16** Elemental mapping images of  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  NSs after OER durability test.



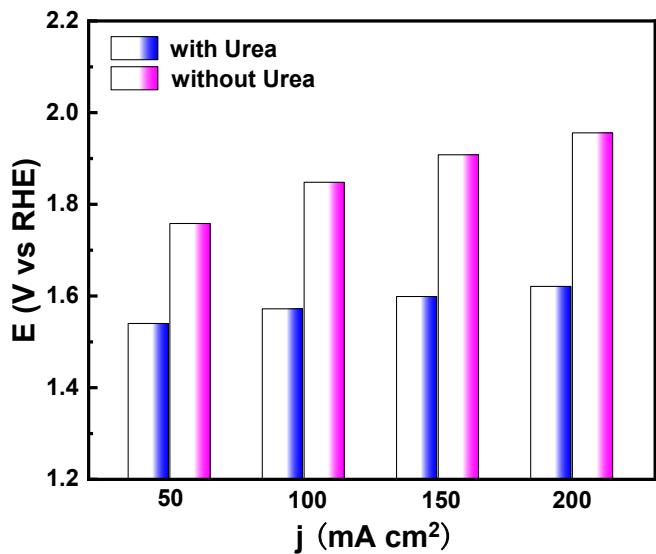
**Fig. S17** Elemental mapping images of  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  NSs after uor durability test.



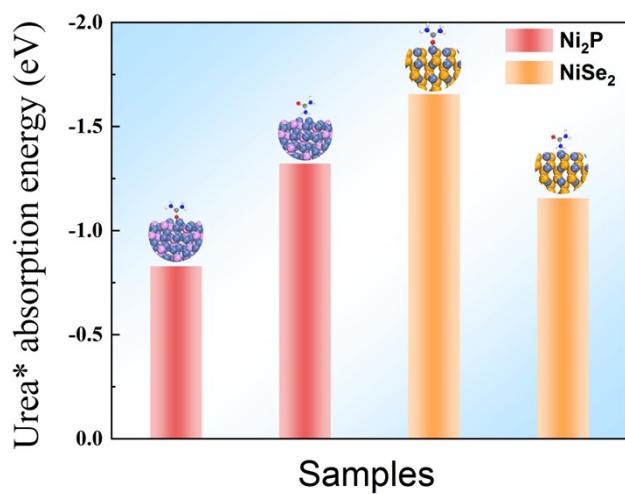
**Fig. S18** TPD adsorption spectra of Ni<sub>2</sub>P/CFC NSs, NiSe<sub>2</sub>/CFC NSs, and Ni<sub>2</sub>P/NiSe<sub>2</sub>/CFC NSs in butylamine/He.



**Fig. S19** TPD adsorption spectra of Ni<sub>2</sub>P/CFC NSs, NiSe<sub>2</sub>/CFC NSs, and Ni<sub>2</sub>P/NiSe<sub>2</sub>/CFC NSs in CO atmospheres.



**Fig. S20** Comparison for cell voltages of  $\text{Ni}_2\text{P}/\text{NiSe}_2/\text{CFC}$  NSs to deliver different current densities for water and urea electrolysis.



**Fig. S21** Comparison of the adsorption energies of  $-\text{NH}_2$  and CO groups in urea molecules adsorbed on  $\text{Ni}_2\text{P}$  and  $\text{NiSe}_2$ .

	<b>0 h</b>	<b>12 h</b>	<b>24 h</b>
<b>pH</b>	<b>13.96</b>	<b>13.91</b>	<b>13.88</b>

**Table S1.** Electrolyte pH changes with long-term stability test time.