Modulating the electronic structure of a hydrogen-bonded organic framework to improve the uranium removal by enhancing hydrogen evolution reaction

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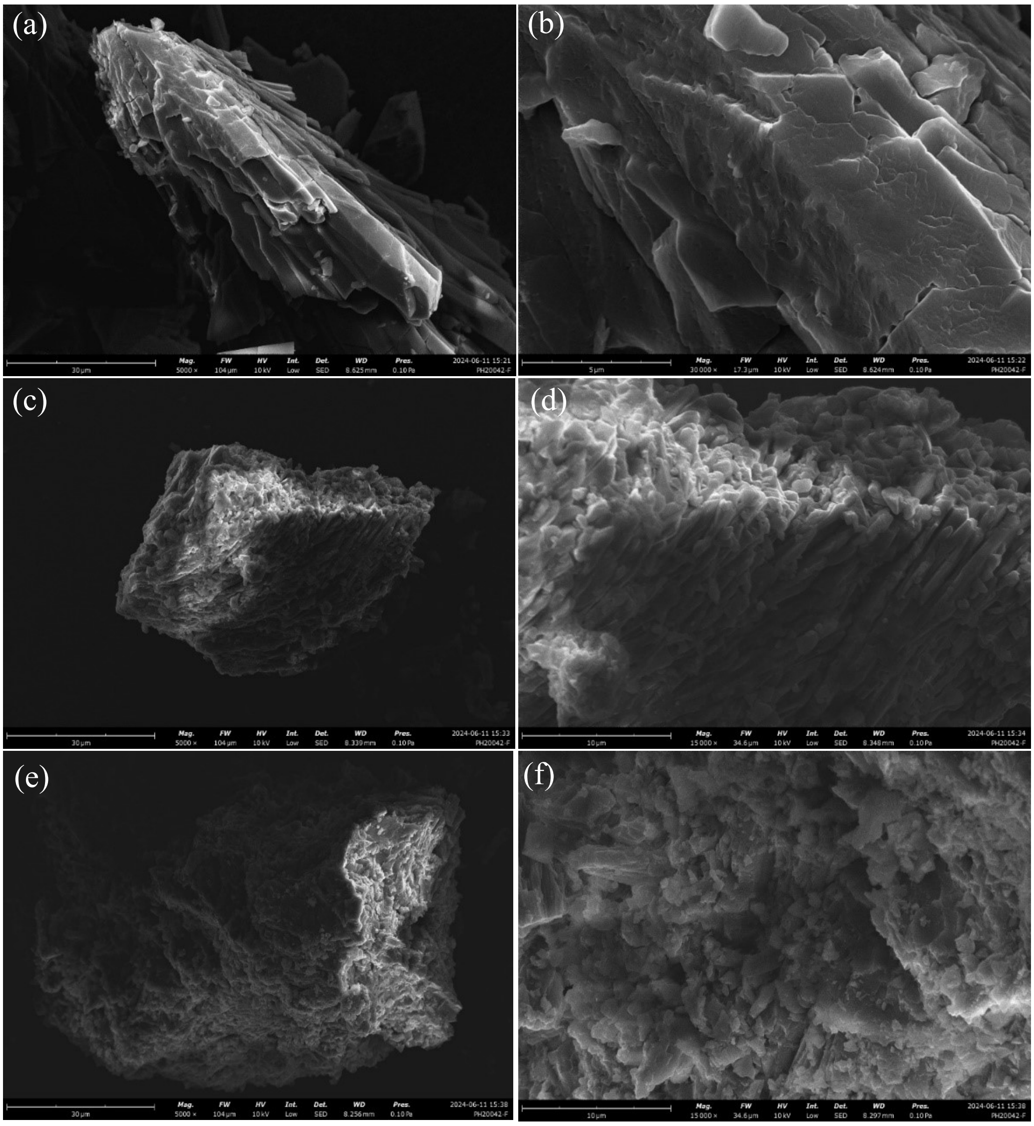

Fig. S1. SEM images of (a, b) Co-HOF; (c,d) Co9.5Ni0.5-HOF; (e, f) Co8Ni2-HOF.

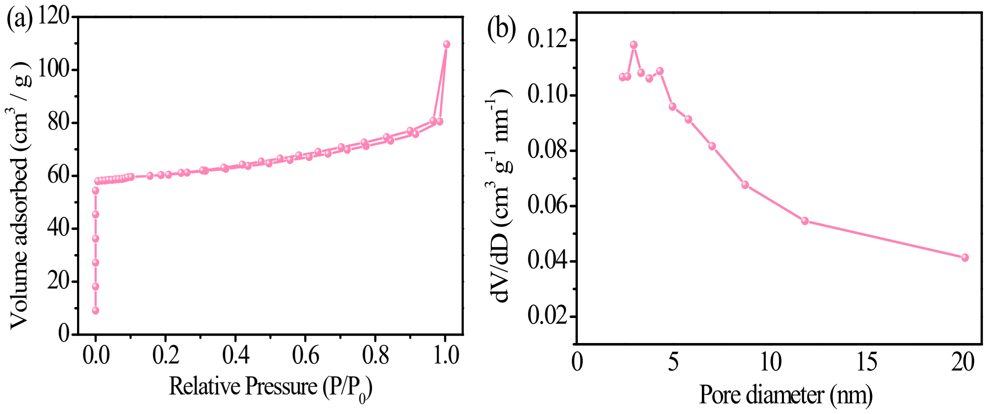

Fig. S2. (a) Nitrogen adsorption-desorption isotherms and (b) pore diameter distribution of Co-HOF.

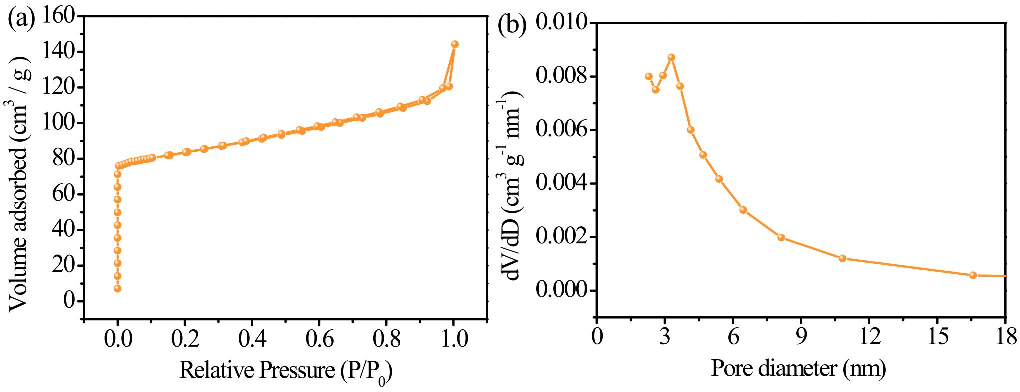

Fig. S3. (a) Nitrogen adsorption-desorption isotherms and (b) pore diameter distribution of $Co_{9.5}Ni_{0.5}$-HOF.

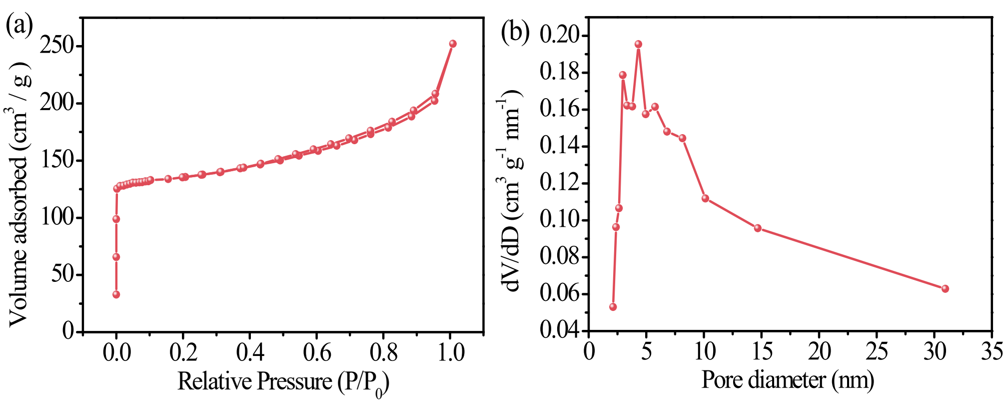

Fig. S4. (a) Nitrogen adsorption-desorption isotherms and (b) pore diameter distribution of Co_9Ni_1-HOF.

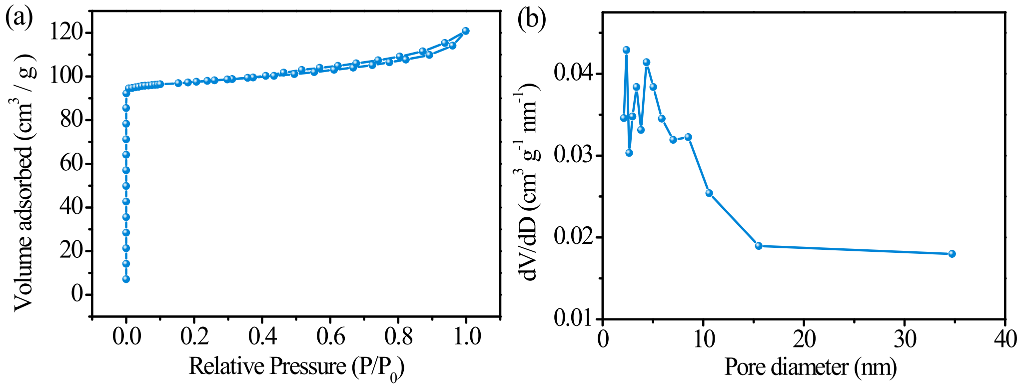

Fig. S5. (a) Nitrogen adsorption-desorption isotherms and (b) pore diameter distribution of Co_8Ni_2-HOF.

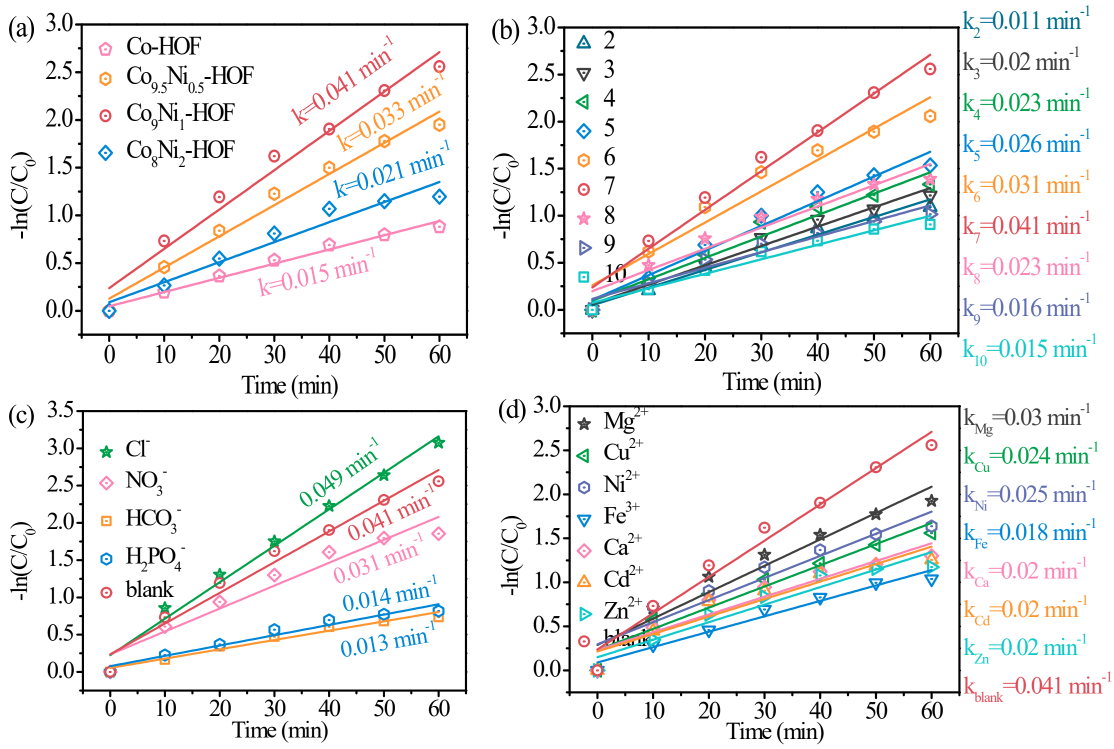

Fig. S6. (a) The kinetic rate constant of the UO_2^{2+} removal over different photocatalysts; (b-d) The kinetic rate constant of the UO_2^{2+} removal for different effects, (b) pH; (c) anions and (d) cations.

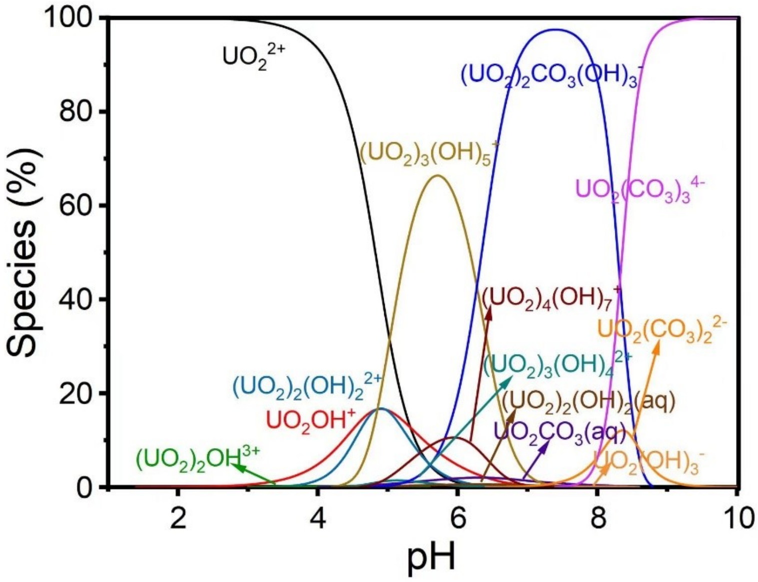

Fig. S7. Simulation of U(VI) speciation as a function with different pH.

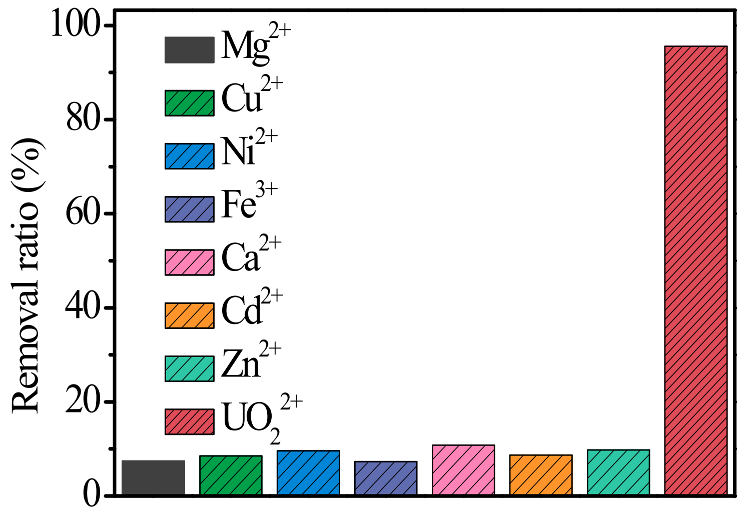

Fig. S8. Selective removal of coexistent ions on Co_9Ni_1-HOF.

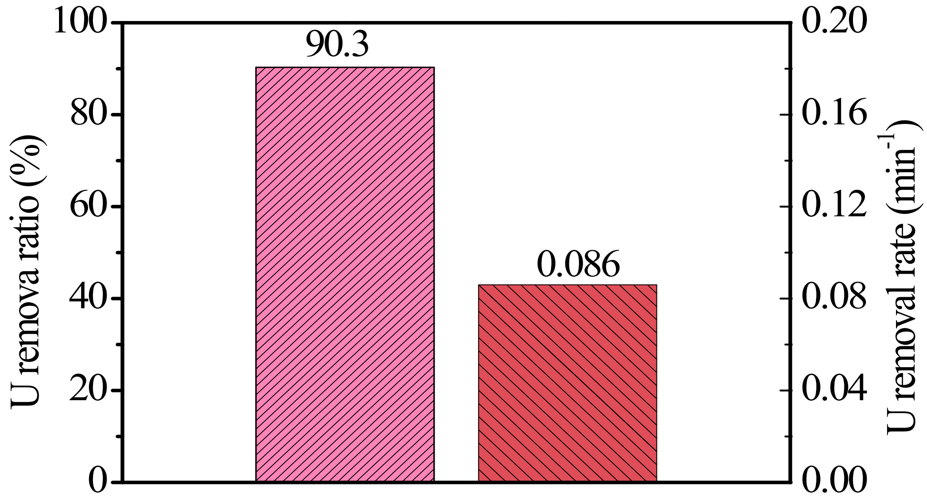

Fig. S9. The removal ratio and rate of U in simulated seawater for Co_9Ni_1-HOF photocatalyst

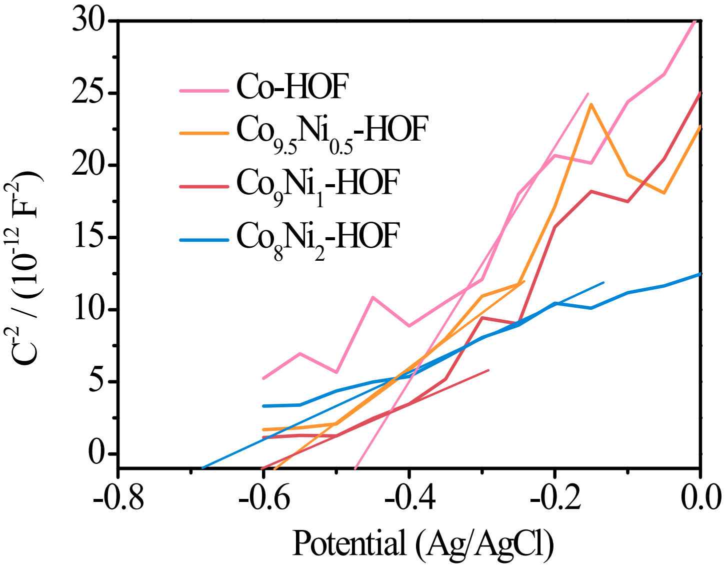

Fig. S10. Mott-Schottky plots of the various samples.

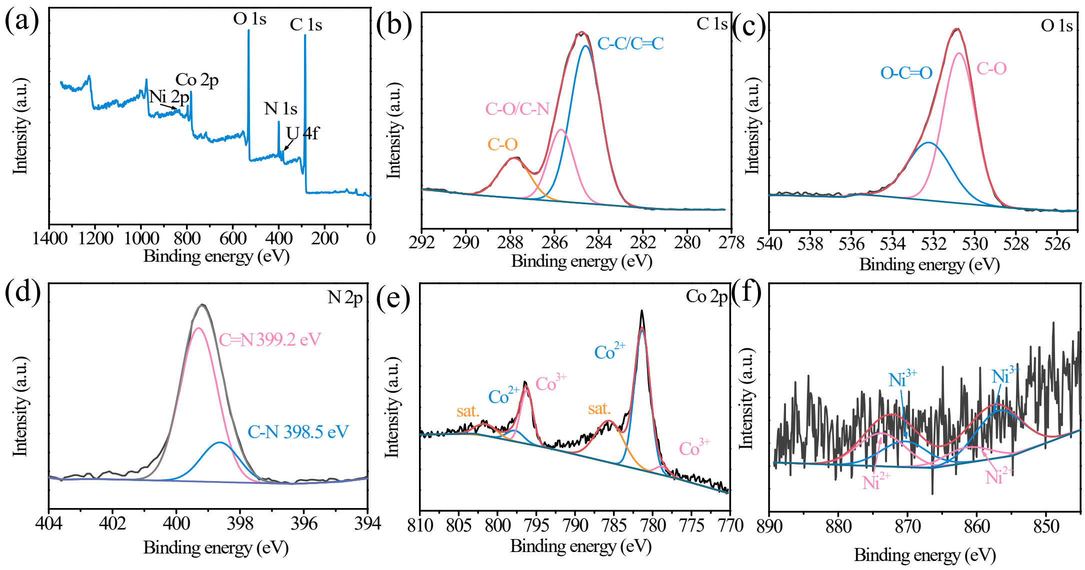

Fig. S11. (a) Full XPS region of Co_9Ni_1-HOF; high-resolution XPS spectra of (b) C 1s, (c) C 1s, (d) N 2p, (e) Co 2p and (d) Ni 2p after uranium removal.

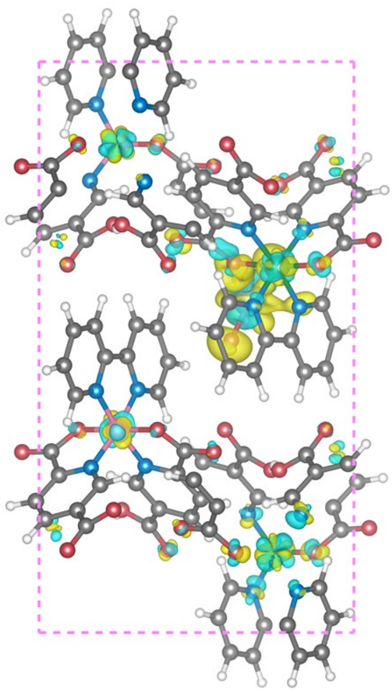

Fig. S12. Charge density differences of UO_2^{2+} adsorption on NiCo-HOF. Yellow indicates electron accumulation, and light blue indicates depletion.

Table S1. Surface and pore information of the samples.

Samples	Specific surface area (m^2/g)	Pore volume (cm^3/g)	Pore diameter (nm)
Co-HOF	191.62	0.137	2.4
$Co_{9.5}Ni_{0.5}$-HOF	308.34	0.192	2.83
Co_9Ni_1-HOF	432.66	0.364	3.67
Co_8Ni_2-HOF	270.85	0.185	2.86

Table S2. The ratio of different valence states over Co^{2+}/Co^{3+} and Ni^{3+}/Ni^{2+}

Samples	$Co^{2+}/Co^{3+}(2p_{1/2})$	$Co^{2+}/Co^{3+}(2p_{3/2})$	$Ni^{3+}/Ni^{2+}(2p_{1/2})$	$Ni^{3+}/Ni^{2+}(2p_{3/2})$
Co-HOF	1.006	1.018	-	-
Co_9Ni_1-HOF	1.008	1.021	1.002	1.003

Table S3. Comparison of HER activity with other materials.

Materials	Overpotential (mV)	Reference
$Mg_{0.99}Ni_{0.01}Ga_{0.01}Fe_{1.99}O_4$	-820	S1
2H-TaS_2	575	S2
Mo-doped SnS	377	S3
Ni-SAO	837.6	S4
Pd@TiO_2-H	430	S5
PSS (BiW@PEPS)	361	S6

B, P and S-doped Ag_2WO_4	330	S7
$Ni/Ni_3C/CdS$	-1080	S8
$Cu-SnO_2$/ZIF-8	364	S9
Cl-doped CuO	400	S10
MoS_2/BN/rGO	-422	S11
CuO Nanoflowers	1020	S12
Ag@g-C_3N_4/r-GO	484	S13
ZnO-Ti_3C_2	495	S14
$MnTiO_3$/g-C_3N_4	357	S15
$Co_{0.6}Cu_{0.4}Fe_2O_4$	-810	S16
Tetra-carboxylic acid based MOF	391	S17
Co_9Ni_1-HOF	355	This work

Reference:

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Table S4. *k* values of the different samples.

Samples	Co-HOF	$Co_{9.5}Ni_{0.5}$-HOF	Co_9Ni_1-HOF	Co_8Ni_2-HOF
k values	0.0027	0.0047	0.0068	0.0031