coatings

Supplementary Materials: **Component Engineering of Multiphase Nickel Sulfide-Based Bifunctional Electrocatalysts for Efficient Overall Water Splitting**

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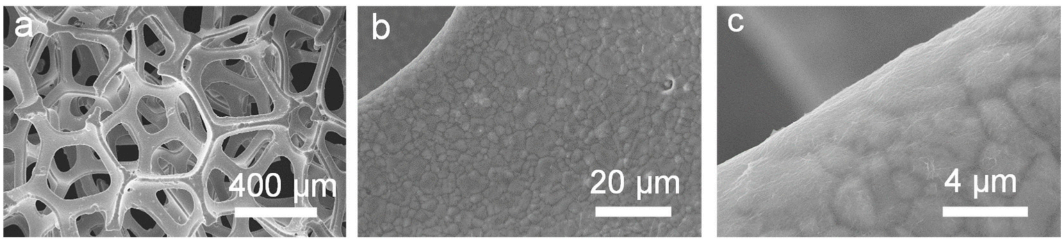

Figure S1. (a-c) The different magnification SEM images of NF surface.

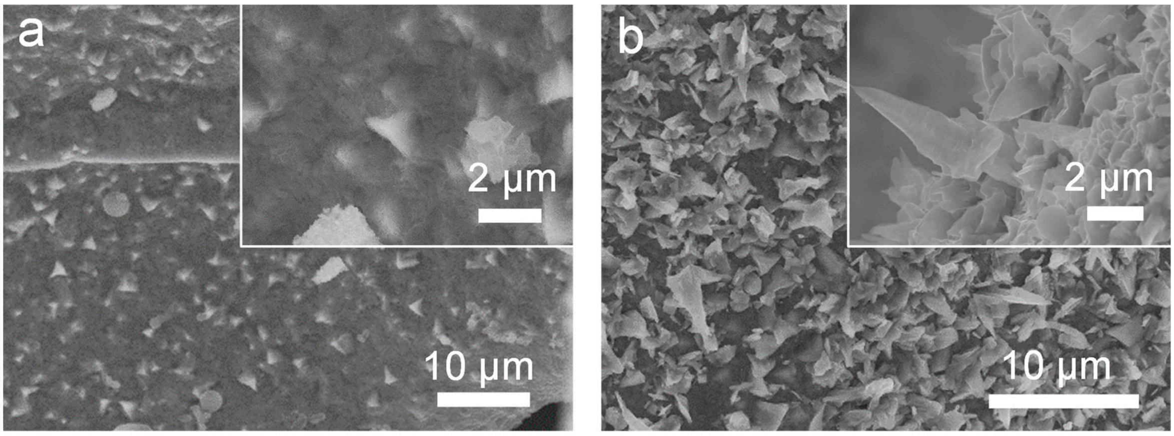

Figure S2. SEM images of Ni_xS_y-2 (a), Ni_xS_y-3 (b). The insets exhibit the corresponding high magnification SEM image, respectively.

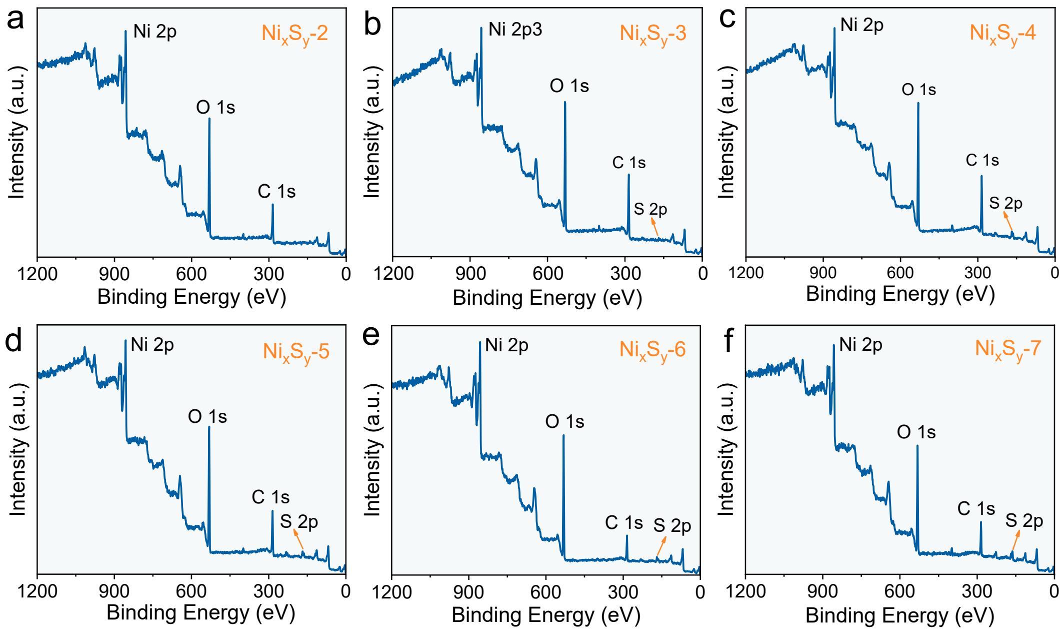

Figure S3. XPS spectra of the survey spectrum for Ni_xS_y-2 (a), Ni_xS_y-3 (b), Ni_xS_y-4 (c), Ni_xS_y-5 (d), Ni_xS_y-6 (e), and Ni_xS_y-7 (f), respectively.

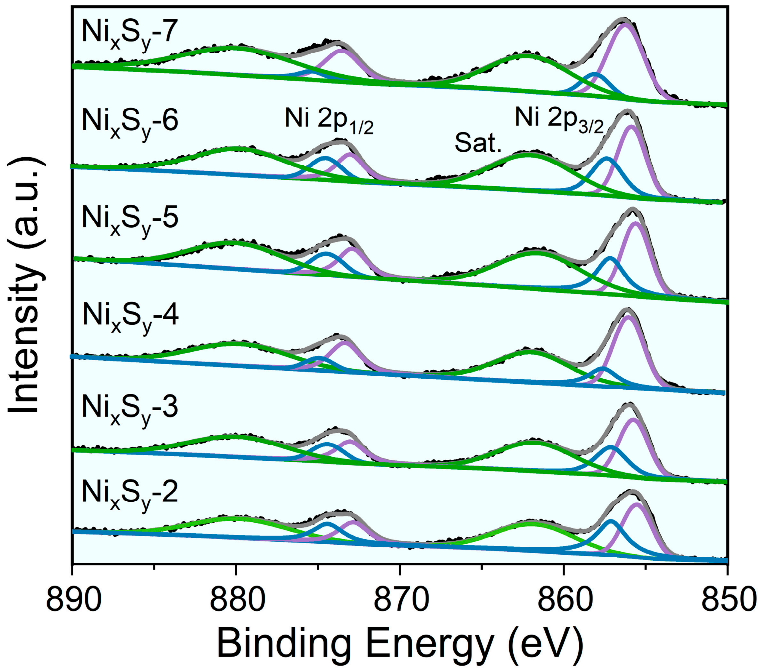

Figure S4. XPS spectra of the Ni 2p spectrum for all the as-prepared samples.

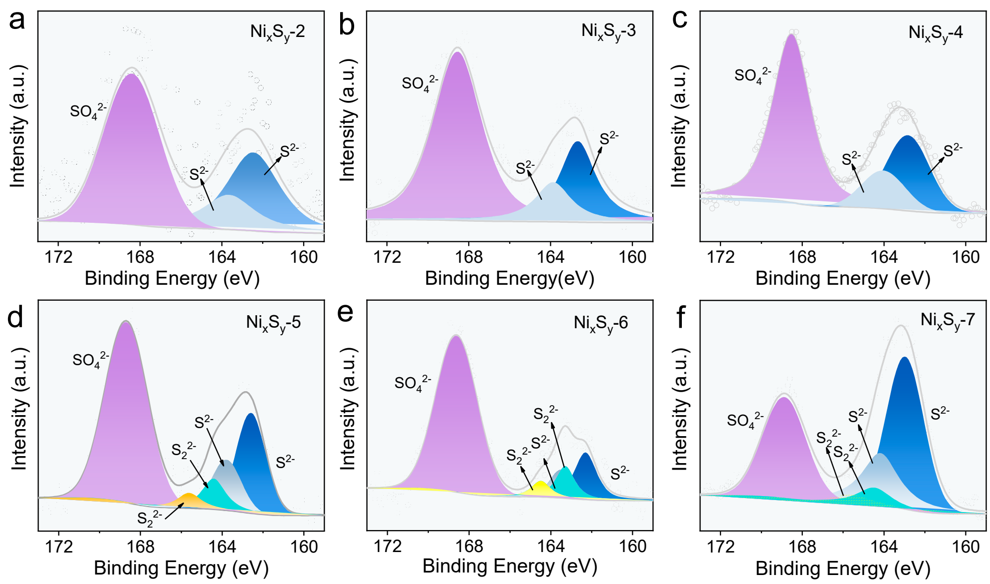

Figure S5. XPS spectra of the S 2p spectrum for Ni$_x$S$_y$-2 (a), Ni$_x$S$_y$-3 (b), Ni$_x$S$_y$-4 (c), Ni$_x$S$_y$-5 (d), Ni$_x$S$_y$-6 (e), and Ni$_x$S$_y$-7 (f), respectively.

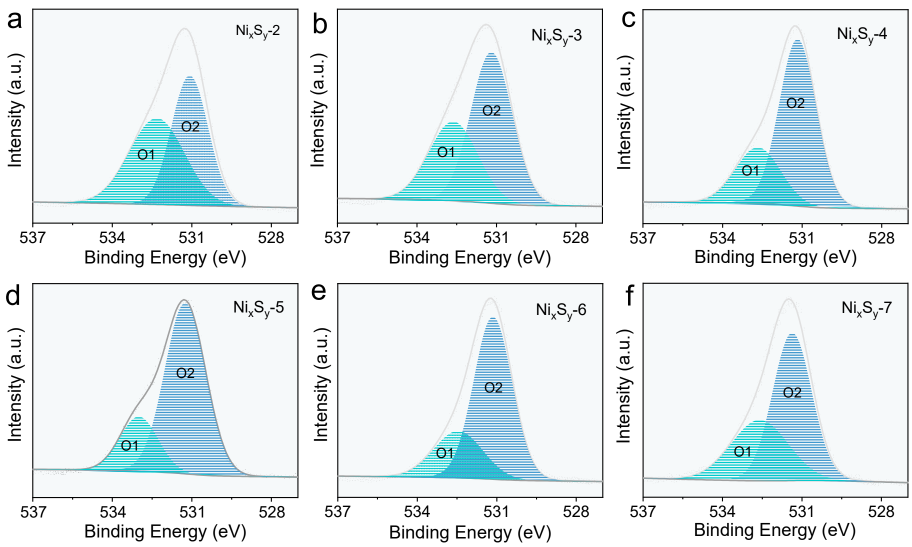

Figure S6. XPS spectra of the O 1s spectrum for Ni_xS_y-2 (a), Ni_xS_y-3 (b), Ni_xS_y-4 (c), Ni_xS_y-5 (d), Ni_xS_y-6 (e), and Ni_xS_y-7 (f), respectively.

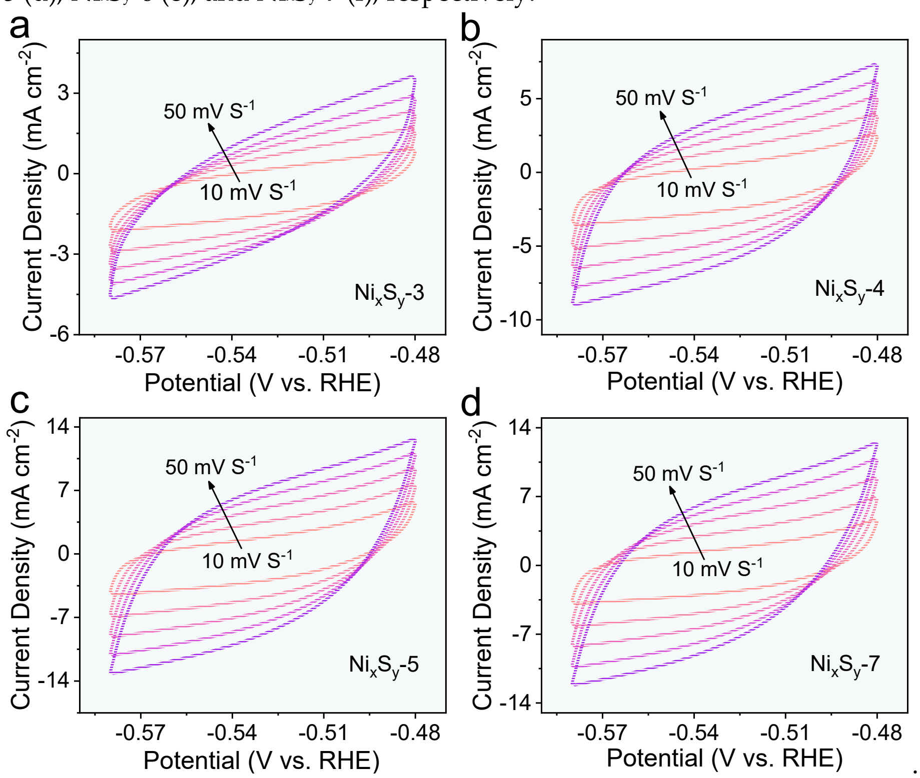

Figure S7. CV curves of the Ni_xS_y-3 (a), Ni_xS_y-4 (b), Ni_xS_y-5 (c), Ni_xS_y-6 (d), respectively, for HER process.

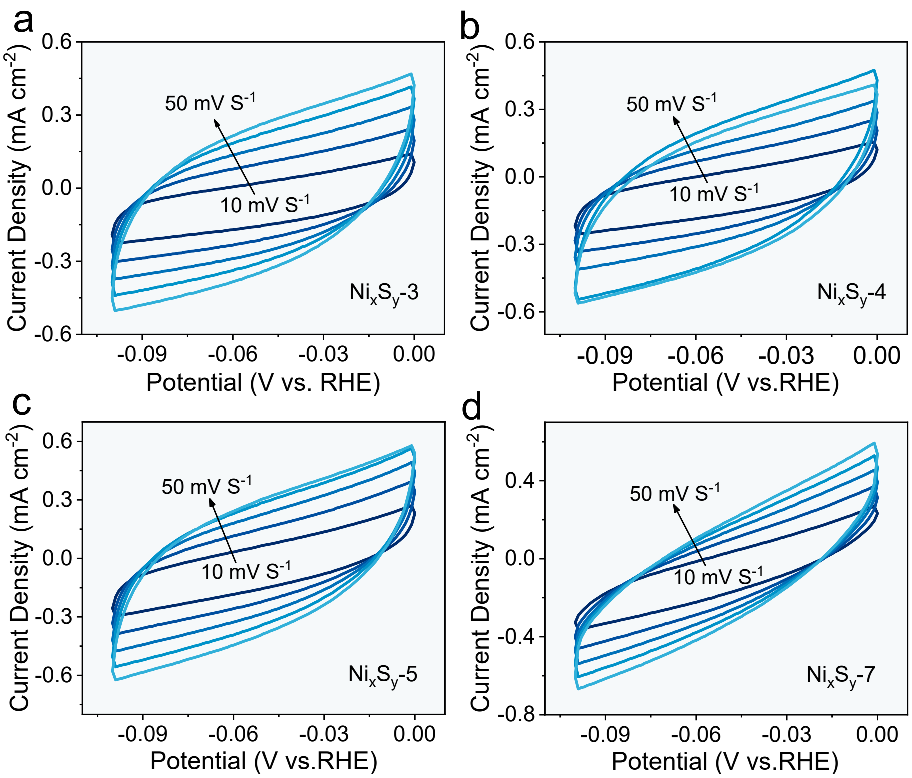

Figure S8. CV curves of the Ni_xS_y-3 (a), Ni_xS_y-4 (b), Ni_xS_y-5 (c), Ni_xS_y-6 (d), respectively, for OER process.

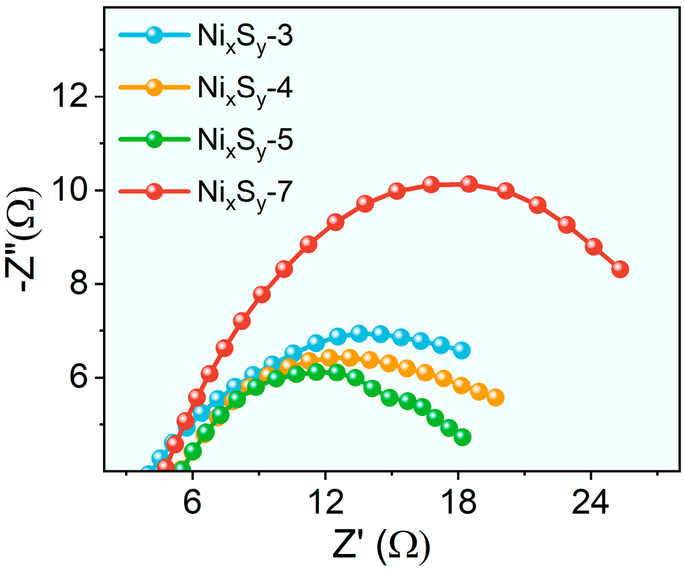

Figure S9. The corresponding EIS plot for Ni_xS_y-3 electrode, Ni_xS_y-4 electrode, Ni_xS_y-5 electrode, and Ni_xS_y-7 electrode.

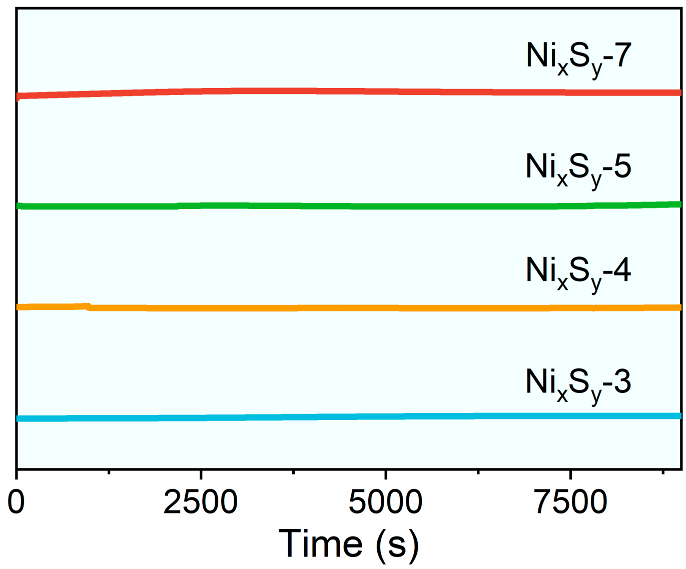

Figure S10. The stability test toward HER at constant current density of 10 mA cm^{-2} for Ni$_x$S$_y$-3 electrode, Ni$_x$S$_y$-4 electrode, Ni$_x$S$_y$-5 electrode, and Ni$_x$S$_y$-7 electrode.

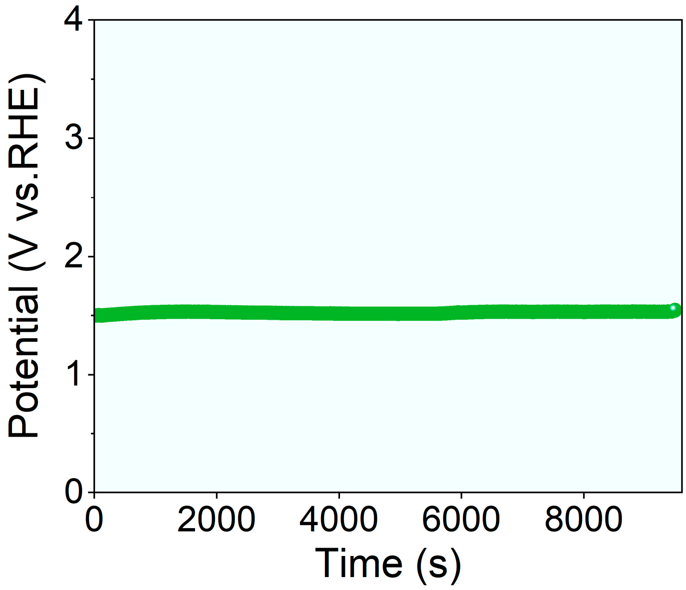

Figure S11. Stability test of overall water splitting at constant current density of 10 mA cm^{-2} for Ni$_x$S$_y$-5 electrolyzer.

Table S1. Summary of the HER activities of recently reported non-noble metal-based electrocatalysts in 1 M KOH.

Catalysts	Tafel (mV dec^{-1})	η_{10} (mV)	Reference
NiCoP	64.4	178	*Adv. Energy Mater.* **2018**, *8*, 1802615
Co_4Mo_2@NC	73.5	218	*J. Mater. Chem. A*, **2017**, *5*, 16929
CoP NFs	56.2	136	*ACS Catal.* **2020**, *10*, 412
Co_3O_4/MoS_2	98	205	*Appl. Catal., B* **2019**, *248*, 202
NiS_2 NA	104	149	*Electrochim. Acta* **2015**, *153*, 508

$NiCo_2S_4$ NW	58.9	210	*Adv. Funct. Mater.* **2016**, *26*, 4661
Mo_2NiB_2	71	160	*Smal,l* **2022**, *18*, 2104303
NiCoP	83	130	*Adv. Energy Mater.* **2018**, *8*, 1800555
$K_2Fe_4O_7$/NF	47.1	109	*J. Mater. Chem. A* **2021**, *9*, 7586
MoB	59	225	*Angew. Chem. Int. Ed.* **2012**, *51*, 12703
Ni_xS_y-5	140	148	This work

Table S2. Summary of the OER activities of recently reported non-noble metal-based electrocatalysts.

Catalysts	**Tafel (mV dec^{-1})**	**η_{10} (mV)**	**Reference**
Mo_2C/NCF	55	144	*ACS Nano* **2016**, *10*, 11337
CeO_x/CoS	50	269	*Angew. Chem. Int., Ed.* **2018**, *57* ,8654
CoNiB	120	302.4	*J. Mater. Chem. A* **2021**, *9*, 6469
Mo_2NiB_2	57	280	*Small* **2022**, *18*, 2104303
Mo-CoOOH	68	305	*Nano Energy*, **2018**, *48*, 73
RuO_2/NF	128	257	*J. Energy Chem.* **2022**, *70*, 472
$K_2Fe_4O_7$/NF	59	308	*J. Mater. Chem. A*, **2021**, *9*, 7586
h-NiSx	96	180	*Adv. Energy Mater.* **2016**, *6*, 1502333
Fe-Ni_3S_2/FeNi	54	282	*Small*, **2017**, *13*, 1604161
$CoNi_2Se_4$	72	160	*Chem. Commun.* **2017**, *53*, 5412
CoOOH/Co_9S_8	86.4	240	*Angew. Chem. Int., Ed.* **2022**, *61*, e2021171
Co/WN-600	65	232	*J. Mater. Chem. A*, **2020**, *8*, 22938
Ag/$Co(OH)_x$	76	250	*Angew. Chem., Int. Ed.* **2020**, *59*, 7245
Ni-MoN	98	276	*Adv. Energy Mater.* **2018**, *8*, 1802327
$Ni_{0.6}Co_{1.4}P$	80	300	*Adv. Funct. Mater.* **2018**, *28*, 1706008
Ni_xS_y-5	60	111	This work

Table S3. Comparison of electrocatalytic performance of Ni_xS_y-5 with recently reported bifunctional electrocatalysts for overall-water-splitting in alkaline media.

Catalysts	**Electrolyte**	**η_{10} (V)**	**Reference**
Pt/C-RuO_2	1 M KOH	1.67 V	*Appl. Catal., B* ***2020**, 278, 119281*
MoS_2/Ni_3S_2	1 M KOH	1.56	*Angew. Chem., Int. Ed.* ***2016**, 55, 6702*

$NiCo_2S_4$/NF	1 M KOH	1.61	*Adv. Mater. Interfaces, 2018,* ***5****, 1701396*
Co_3O_4/MoS_2	1 M KOH	205	*Appl. Catal. B-Environ.* **2019**, *248*, 202
Mo-CoOOH	1 M KOH	1.56	*Nano Energy,* ***2018****, 48, 73*
CoP NFs	1 M KOH	1.65	*ACS Catal.* **2020**, *10*, 412
Co_3O_4 NCs	1 M KOH	1.91	*Chem. Commun.* ***2015****, 51, 8066*
$K_2Fe_4O_7$/NF	1 M KOH	1.59	*J. Mater. Chem. A* ***2021****, 9, 7586*
Mo_2NiB_2	1 M KOH	1.57	*Small* **2022**, *18*, 2104303
$NiCo_2S_4$	1 M KOH	1.58	*Adv. Funct. Mater.* ***2019****, 29, 1807031*
Ni_xS_y-5	1 M KOH	1.48	This work