Electronic Supplementary Material (ESI) for Inorganic Chemistry Frontiers.

Supporting Information

Anchoring stable FeS_2 nanoparticles on MXene nanosheets via interface engineering for efficient water splitting

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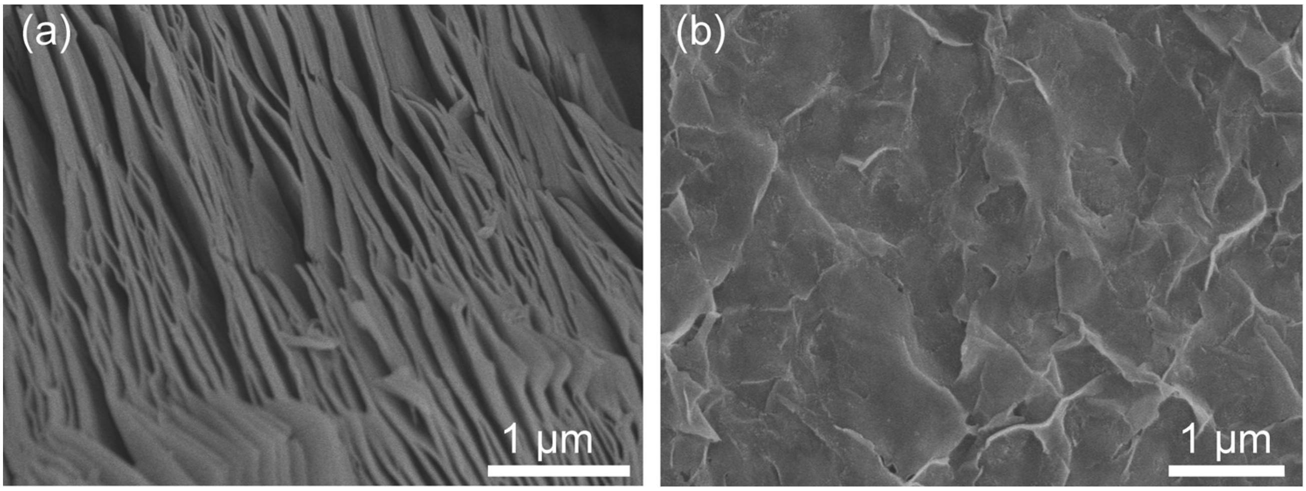

Fig. S1. SEM images of (a) accordion-like $Ti_3C_2T_x$ MXene and (b) exfoliated ultrathin MXene nanosheets, respectively.

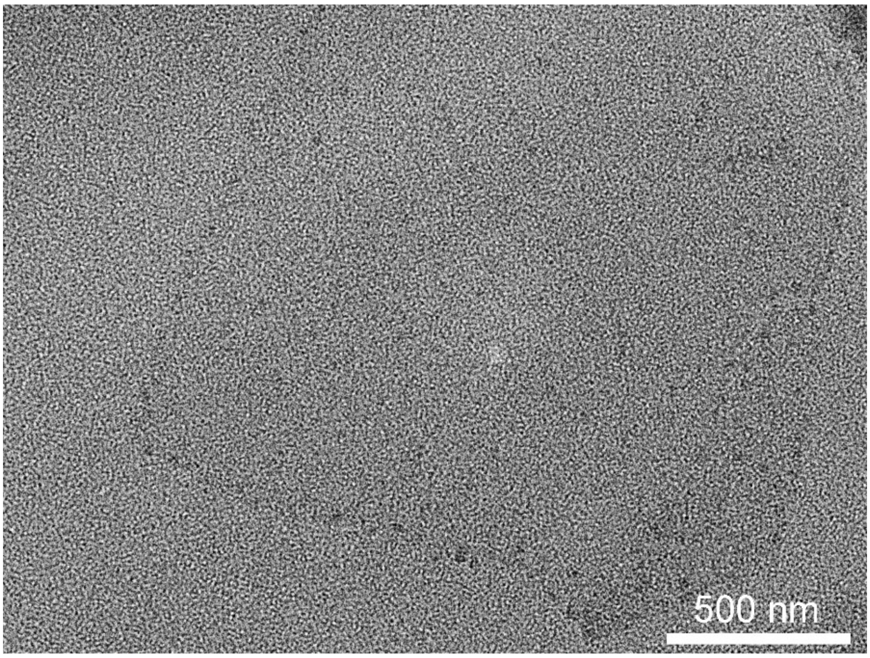

Fig. S2. TEM image of exfoliated MXene.

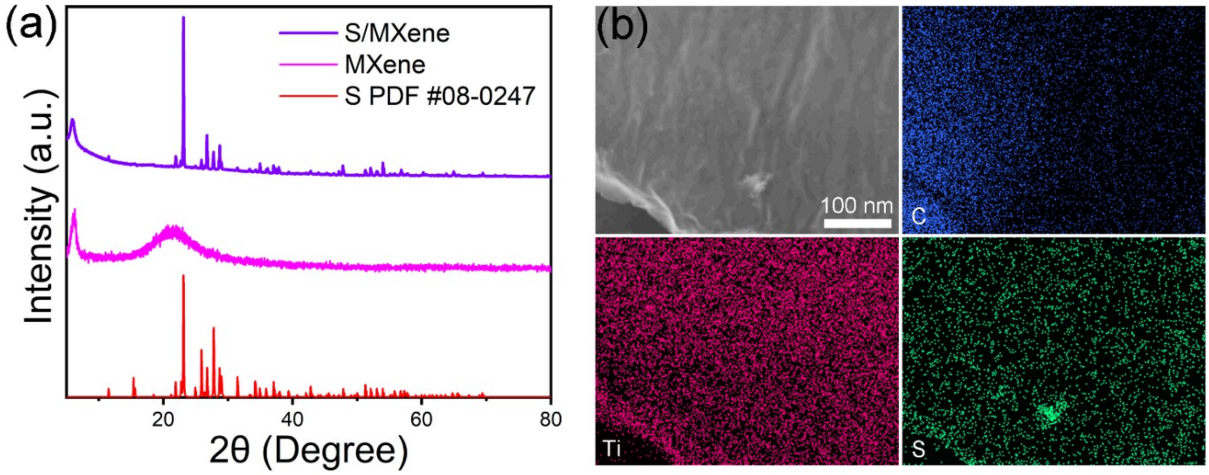

Fig. S3. (a) XRD patterns and (b) SEM mapping of S/MXene mixture.

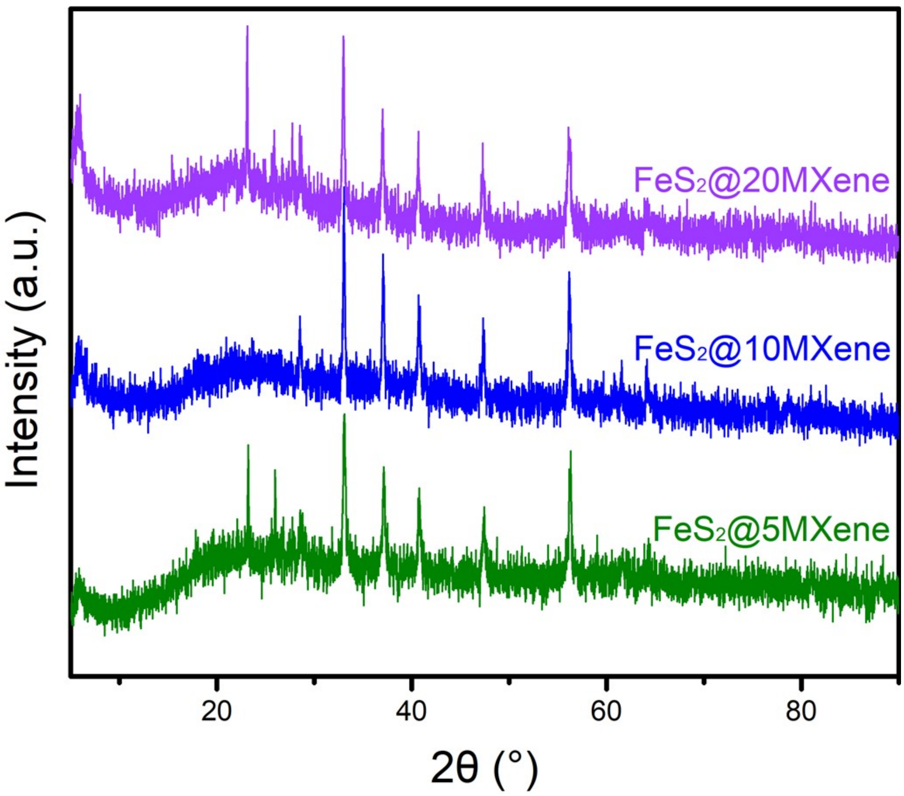

Fig. S4. XRD patterns of FeS_2@5MXene, FeS_2@10MXene and FeS_2@20MXene.

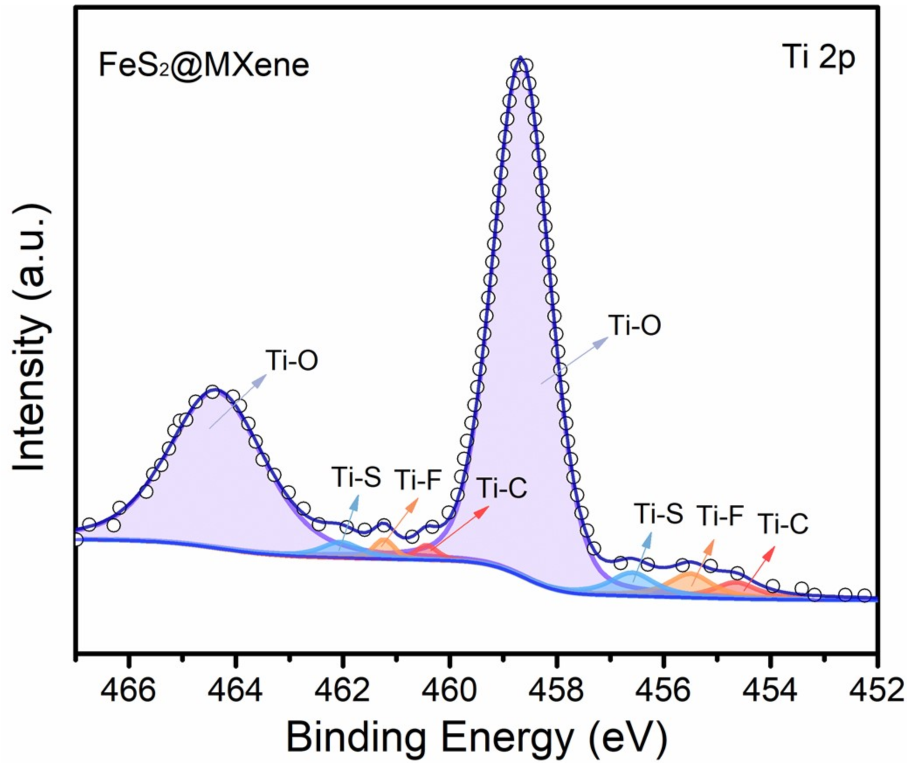

Fig. S5. Ti 2p spectrum of FeS_2@MXene.

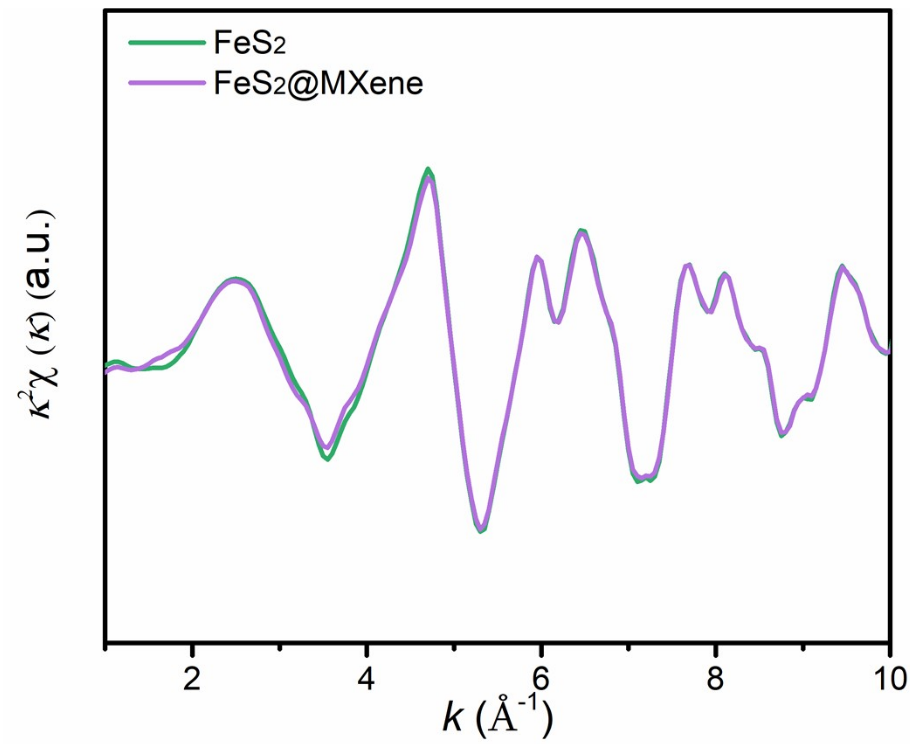

Fig. S6. Fe K-edge XAFS $k^2[\chi(k)]$ oscillation curves.

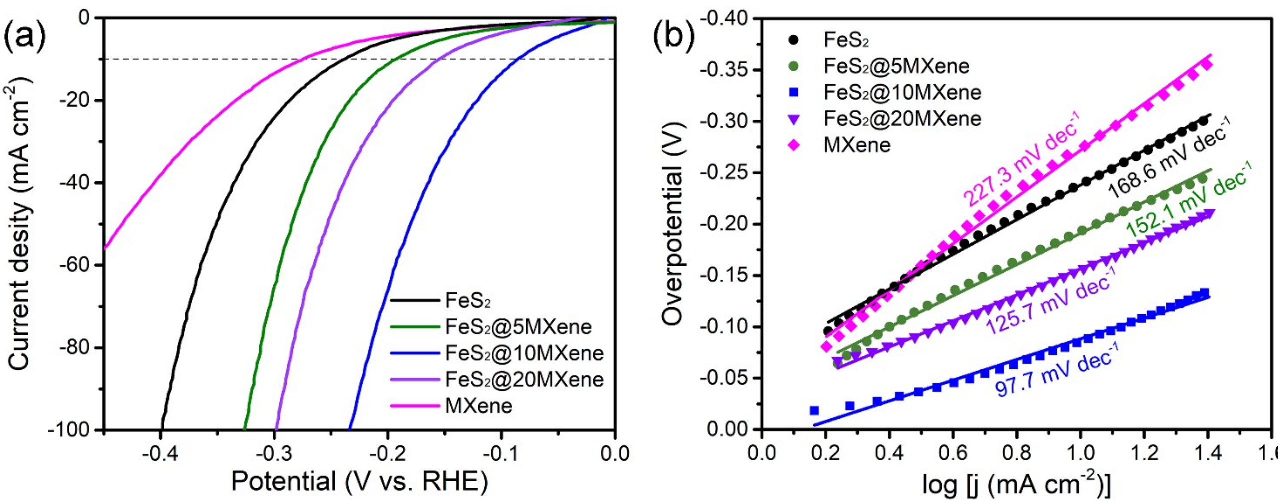

Fig. S7. HER performance in 1 M KOH. (a) LSV curves, (b) Tafel plots of FeS_2, FeS_2@5MXene, FeS_2@10MXene, FeS_2@20MXene and MXene.

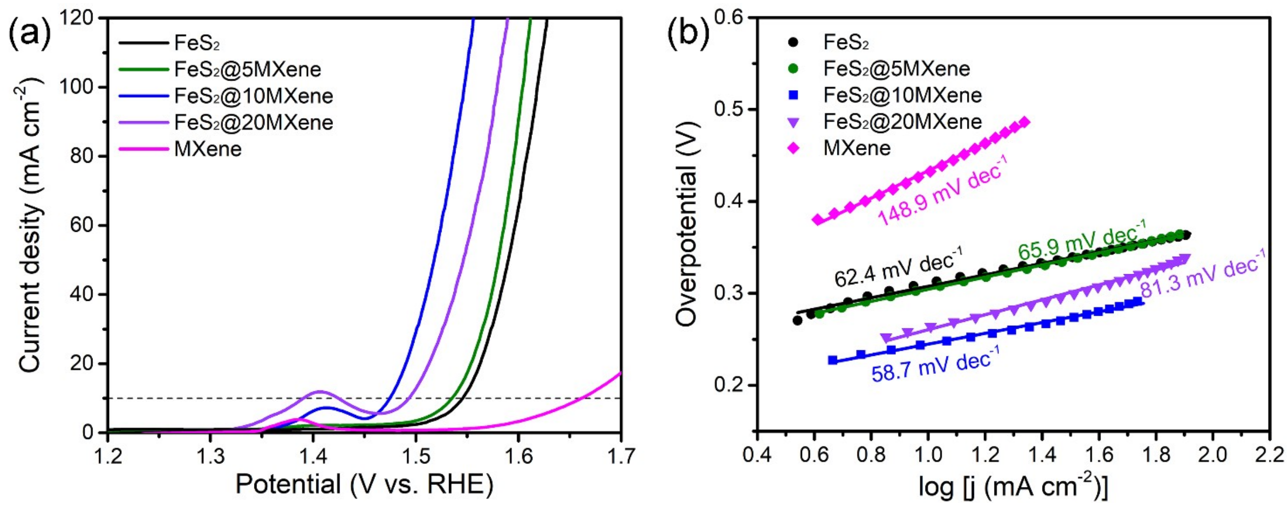

Fig. S8. OER performance in 1 M KOH. (a) LSV curves, (b) Tafel plots of FeS_2, FeS_2@5MXene, FeS_2@10MXene, FeS_2@20MXene and MXene.

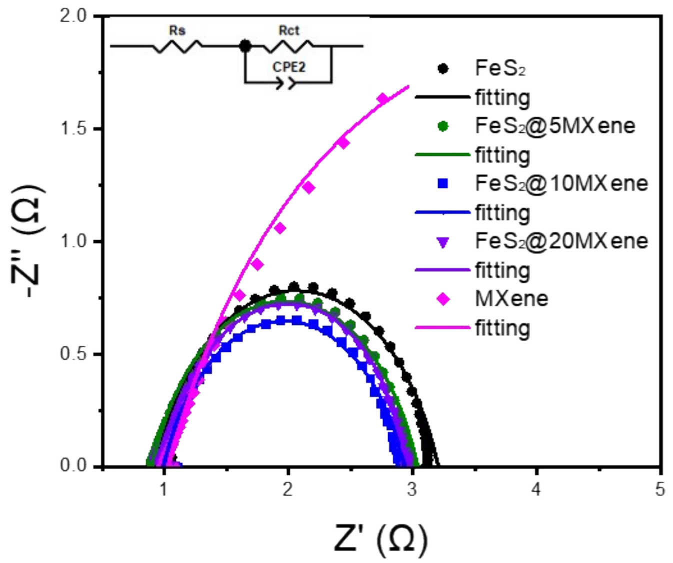

Fig. S9. Nyquist plots of FeS_2, FeS_2@5MXene, FeS_2@10MXene, FeS_2@20MXene and MXene.

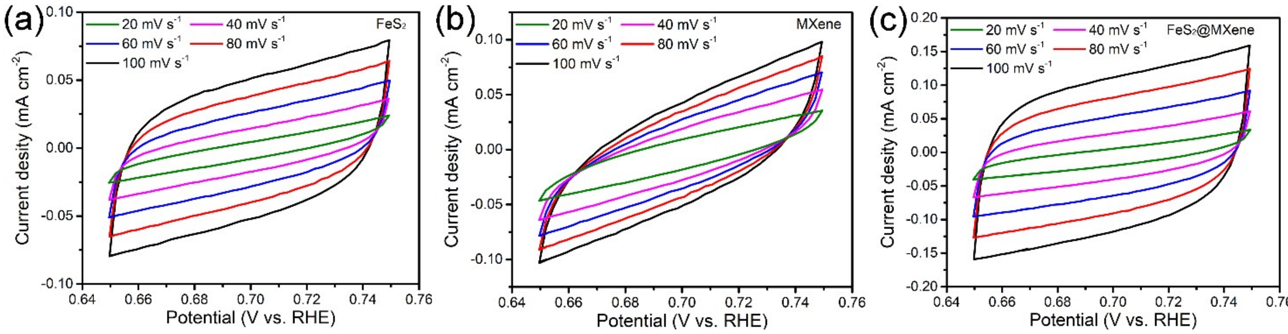

Fig. S10. Cyclic voltammograms of (a) FeS_2, (b) MXene and (c) FeS_2@MXene in 1.0 M KOH at various scanning rates (from 20 mV to 100 mV s^{-1}), within a potential range where no Faradaic process is observed.

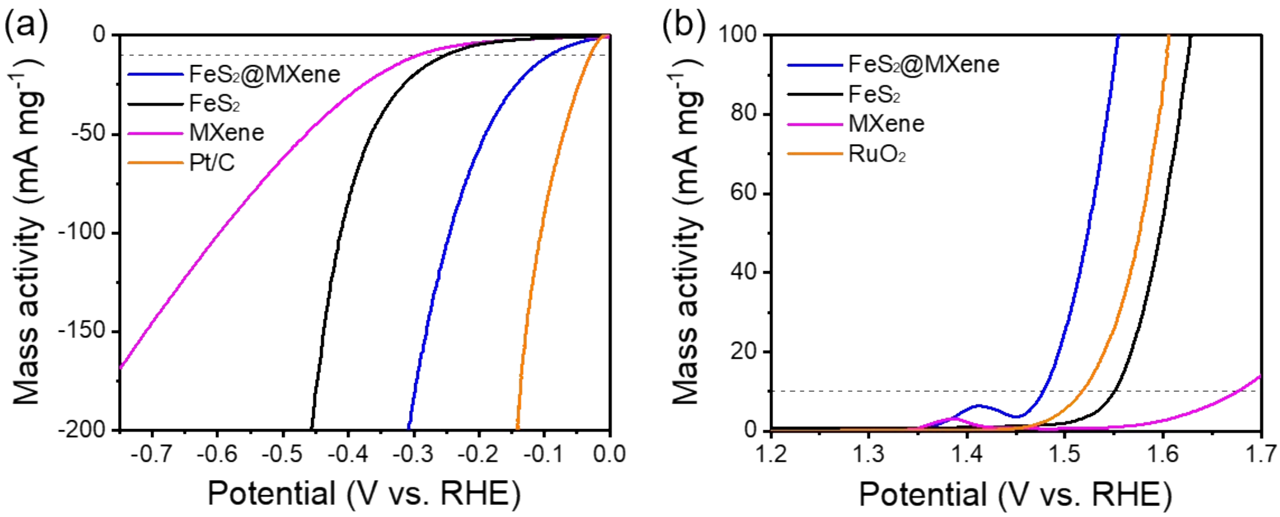

Fig. S11. (a) HER and (b) OER polarization curve normalized by the loading mass for MXene, FeS_2 and FeS_2@MXene.

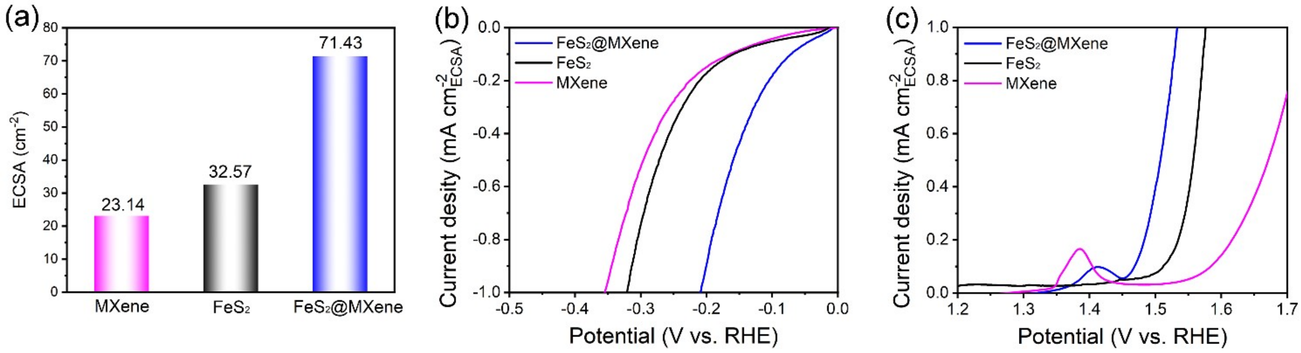

Fig. S12. (a) The ECSA of MXene, FeS_2 and FeS_2@MXene. (b) HER and (c) OER polarization curve normalized by the ECSA for MXene, FeS_2 and FeS_2@MXene.

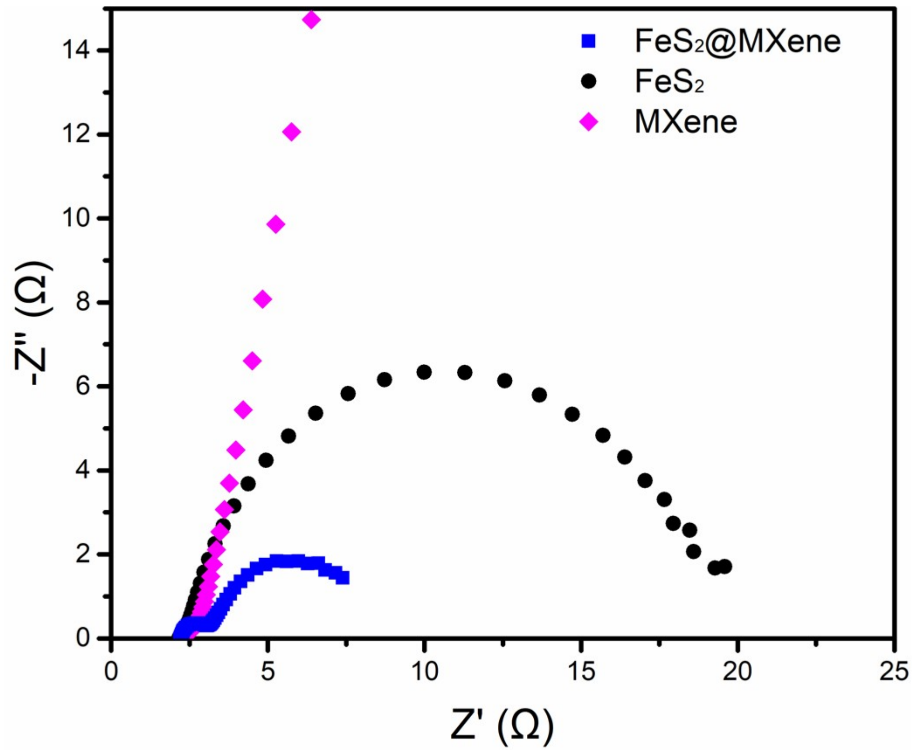

Fig. S13. Nyquist curves of FeS_2, FeS_2@10MXene and MXene towards water splitting.

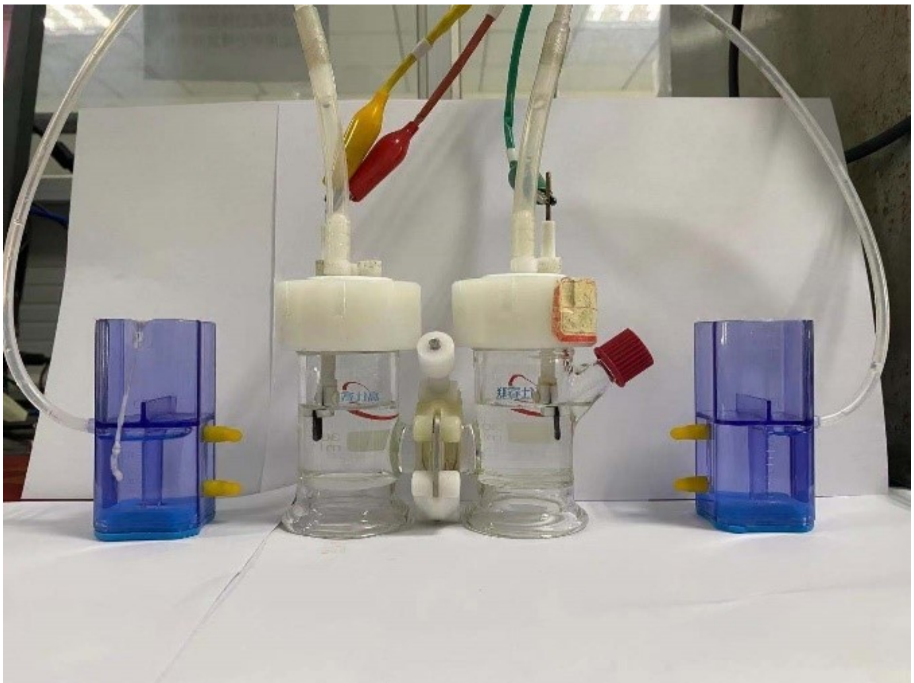

Fig. S14. Digital photograph of the H-type electrolyzer for Faraday efficiency test.

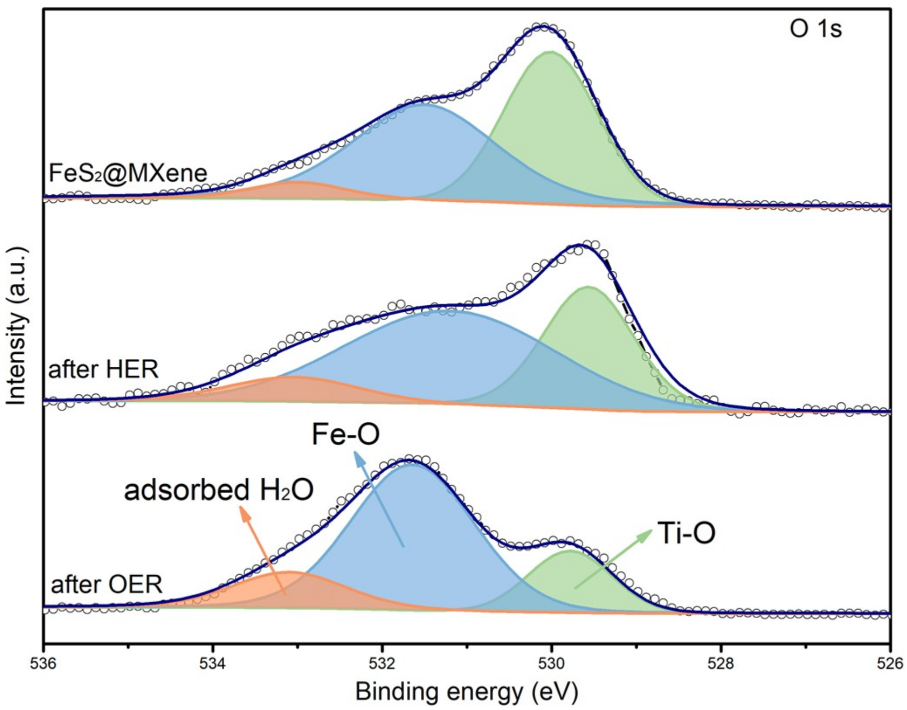

Fig. S15. O 1s spectrum of FeS_2@MXene before and after HER/OER test.

Table S1. HER Fitting results of EIS for MXene, FeS_2 and FeS_2@MXene.

Catalysts	Rs (ohm)	Rct (ohm)	R1
MXene	1.01	15.11	0.50
FeS_2	1.03	6.52	1.16
FeS_2@10MXene	0.91	4.99	0.424

Table S2. OER Fitting results of EIS for MXene, FeS_2 and FeS_2@MXene.

Catalysts	Rs (ohm)	Rct (ohm)
MXene	1.03	5.85
FeS_2	0.92	2.29
FeS_2@5MXene	0.99	1.94
FeS_2@10MXene	0.99	1.94
FeS_2@20MXene	0.94	2.05

Table S3. Comparison of the **HER** electrocatalytic activity of FeS_2@MXene in 1.0 M KOH with some representative catalysts based on transition-metal-sulfide-based electrocatalysts which have recently reported.

Sample	Tafel slope (mV dec^{-1})	Current density (mA cm^{-2})	Overpotential (mV)	References
FeS_2@MXene	97.7	10	87	This work
FeS_2/C nanoparticles	98	10	202	Dalton Trans., 2018, 47, 14917-14923
FeS_2/CoS_2 NSs	44	10	78.2	Small 2018, 14, 1801070
NiS_2/FeS_2/NC	115.6	10	172	J. Power Sources, 2019, 436, 226857
$Ni_{0.7}Fe_{0.3}S_2$	109	10	155	J. Mater. Chem. A, 2017, 5, 15838-15844
Fe11.1%-Ni_3S_2/Ni foam	89	10	203	J. Mater. Chem. A, 2018, 6, 4346-4353
Ni-FeS_2-0.5	-	10	164	Inorg. Chem. 2019, 58, 7615-7627
FS-0.9-30	112.51	10	157	ACS Sustainable Chem. Eng. 2019, 7, 18015-18026
Mesoporous FeS_2	78	10	96	J. Am. Chem. Soc. 2017, 139, 13604-13607
P-(Ni, Fe)$_3S_2$/NF	88	10	98	ACS Appl. Mater. Interfaces 2019, 11, 27667-27676

$Co_{0.25}Fe_{0.75}S_2$	58	10	267	New J. Chem., 2020, 44, 1711-1718
FeS_2@C	128	10	195	ACS Appl. Nano Mater. 2019, 2, 3889-3896
$Ni_{0.9}Fe_{0.1}PS_3$@MXene	114	10	196	Adv. Energy Mater. 2018, 8, 1801127
Co-MoS_2/Mo_2CTx	82	10	112	Nanoscale, 2019, 11, 10992-11000
MoS_2/ Ti_3C_2-MXene@C	45	10	135	Adv. Mater., 2017, 29, 1607017
NiS_2/V-MXene	85	10	179	J. Catal., 2019, 375, 8-20
Ni_3S_4/NiS_2/FeS_2	110	10	197	Appl. Surf. Sci., 2021, 560, 149985
Co_9S_8-MoS_2/NF,	30.5	10	110	Adv. Funct. Mater., 2020, 30, 2002536
Mo-Ni_3S_2 QDs	67	10	115	Nanoscale, 2021, 13, 6644-6653
Fe-c-Ni_3S_2/NF	105	10	115	Dalton Trans., 2021, 50, 7776-7782
FeS_2/PANI /EG	144	10	230	Appl. Surf. Sci. 2020,505, 144534
P-Ni_3S_2/NF	88	10	101	Catal. Sci. Technol., 2020, 10, 7581-7590
NiFe-MS/MOF@NF	82	10	90	Adv. Sci. 2020, 7, 2001965
$Fe_xNi_{3-x}S_2$@NF	71	10	72	Adv. Energy Mater. 2020, 10, 2001963
$NiCo_2S_4$/Ni_3S_2	105	10	119	ACS Appl. Mater. Interfaces 2018, 10, 10890.

$NiCo_2S_4$/CC	82.5	10	150	Catal. Sci. Technol., 2020, 10, 1056-1065
Al-Ni_3S_2/NF	75	10	86	Nanoscale, 2020, 12, 24244-24250
Mo-NiP_x/NiS_y	49	10	137	Adv. Funct. Mater. 2021, 31, 2101532
Fe-doped Co-Mo-S/NF	50.3	10	105	Dalton Trans., 2020, 49, 15009-15022
Mo-doped Ni_3S_2/NF	131.7	10	97.1	ChemistrySelect 2019, 4, 12328.
Ni_3S_2-NTFs	75.7	10	135	Appl. Catal. B, 2019, 243, 693–702
NF/Ni_3S_2-NiOOH	92	10	160	Appl. Surf. Sci., 2018, 456 (31), 164
Ni_3S_2@G	66	10	103	Appl. Surf. Sci., 2019, 465, 772
MoS_2/Ni_3S_2	61	10	98	ACS Catal., 2017, 7, 2357-2366.
Co_3S_4@MoS_2	43	10	136	Nano Energy, 2018, 47, 494-502.

Table S4. Comparison of the **OER** electrocatalytic activity of FeS_2@MXene in 1.0 M KOH with some representative catalysts based on transition-metal-sulfide-based electrocatalysts which have recently reported.

Sample	Tafel slope (mV dec^{-1})	Current density (mA cm^{-2})	Overpotential (mV)	References
FeS_2@MXene	58.7	10	240	This work
FeS_2/C nanoparticles	92	10	240	Dalton Trans., 2018, 47, 14917-14923
FeS_2/C Nanowires	65.6	10	291	Materials, 2019, 12, 3364
FeS_2/CoS_2 NSs	44	100	302	Small 2018, 14, 1801070
$Co_{0.25}Fe_{0.75}S_2$	50	10	324	New J. Chem., 2020, 44, 1711-1718
Co-FeS_2/CoS_2	73	10	278	RSC Adv., 2018, 8, 28684-28691
Ni-FeS_2@Ni-FeOOH	42.6	10	230	J. Mater. Chem. A, 2017, 5, 4335-4342
Ni3S_4/NiS_2/FeS_2	49	10	230	Appl. Sur. Sci. 2021, 560, 149985
$Fe_{11.1\%}$-Ni_3S_2/Ni foam	61.7	10	252	J. Mater. Chem. A, 2018, 6, 4346-4353
Ni-FeS_2-0.5	34	10	250	Inorg. Chem. 2019, 58, 7615-7627
$Co_{0.25}Fe_{0.75}S_2$	50	10	324	New J. Chem., 2020, 44, 1711-1718

C-coated CoS_2-FeS_2	105	10	240	Dalton Trans., 2020, 49, 13352-13358
$NiCoS/Ti_3C_2T_x$	58	10	365	ACS Appl. Mater. Interfaces 2018, 10, 22311-22319
$Ni_{0.7}Fe_{0.3}PS_3$@Mxene	36.5	10	282	Adv. Energy Mater. 2018, 8, 1801127
$Ni_3S_4/NiS_2/FeS_2$	49	10	230	Appl. Surf. Sci. 2021, 560,149985
S-$NiFe_2O_4$@ Ti_3C_2@NF	46.8	20	270	Electrochim. Acta, 2019, 296, 762-770
$Fe_xNi_{1-x}S_2$/C	43	10	248	Renew. Energ., 2021, 168, 416-423
FeS_2/PANI /EG	98	10	260	Appl. Sur. Sci. 2020, 505, 144534
P-Ni_3S_2/NF	30	10	256	Catal. Sci. Technol., 2020, 10, 7581-7590
Fc-Co_xS_y	54.2	10	304	Small 2020, 16, 2001665
Fe-doped Co-Mo-S/NF	79.3	10	268	Dalton Trans., 2020, 49, 15009-15022
Co_3S_4@MoS_2	74	10	280	Nano Energy, 2018, 47, 494-502.
MoS_2/NiS_2 nanosheets	92	10	278	Adv. Sci., 2019, 6, 1900246.
Fe-MoS_2/Ni_3S_2	60	10	256	Dalton Trans., 2019, 48, 12186-12192.

Table S5. Comparison of the **water splitting** electrocatalytic activity of FeS_2@MXene in 1.0 M KOH with some representative catalysts based on transition-metal-sulfide-based electrocatalysts which have recently reported.

Sample	Current density (mA cm^{-2})	Potential (V)	References
FeS_2@MXene	10	1.57	This work
FeS_2/C nanoparticles	10	1.72	Dalton Trans., 2018, 47, 14917-14923
$Ni_{0.7}Fe_{0.3}S_2$	10	1.63	J. Mater. Chem. A, 2017, 5, 15838-15844
Fe11.1%-Ni_3S_2/Ni foam	10	1.60	J. Mater. Chem. A, 2018, 6, 4346-4353
NiS_2/FeS_2/NC	10	1.58	J. Power Sources, 2019, 436, 226857
$Co_{0.25}Fe_{0.75}S_2$	10	1.60	New J. Chem., 2020, 44, 1711--1718
C-coated CoS_2-FeS_2	10	1.66	Dalton Trans., 2020, 49, 13352-13358
Ni_3S_4/NiS_2/FeS_2	10	1.68	Appl. Sur. Sci., 2021, 560, 149985
Fe-doped Ni_3S_2-NF	10	1.56	Nanoscale, 2021, 13, 1807-1812
MoS_2-Ni_3S_2/NF	10	1.56	Inorg. Chem. Front., 2020, 7, 3588-3597
NiFe-MS/MOF@NF	10	1.61	Adv. Sci. 2020, 7, 2001965

Fe-doped Co-Mo-S/NF	10	1.61	Dalton Trans., 2020, 49, 15009-15022
Ni-FeS_2-0.5	10	1.55	Inorg. Chem. 2019, 58, 11, 7615-7627
$Ni_{0.7}Fe_{0.3}PS_3$@MXene || $Ni_{0.9}Fe0_{.1}PS_3$@MXene	10	1.65	Adv. Energy Mater. 2018, 8, 1801127
Ni_3S_2-NGQDs/NF	10	1.58	Small 2017, 13, 1700264.
MoS_2/NiS nanosheets	10	1.61	J. Mater. Chem. A, 2018, 6, 9833-9838.
Fe-MoS_2/Ni_3S_2	10	1.61	Dalton Trans., 2019, 48, 12186-12192.
$Ni_{0.7}Fe_{0.3}S_2$	10	1.62	J. Mater. Chem. A, 2017, 5, 15838-15844.
N-$NiMoO_4$/NiS_2	10	1.6	Adv. Funct. Mater., 2019, 29, 1805298.
NiS_2/CoS_2	10	1.61	Chem. Commun., 2019, 55, 3781-3784.
Co_9S_8/Ni_3S_2	10	1.64	Appl. Catal. B Environ., 2019, 253, 246-252.
Ni_3S_2/NF	10	1.59	J. Energy Chem., 2017, 26, 1217-1222.
Ni_3S_2 nanorods	10	1.61	J. Energy Chem., 2020, 46, 178-186.
NiS film	10	1.64	Chem. Commun., 2016, 52, 1486-1489.
Co-FeS_2 nanospheres	10	1.6	New J. Chem., 2020, 44, 1711-1718.

FeS/NiS/Ni foam	10	1.62	Electrochim. Acta, 2020, 332, 135534.
CoS_2 nanosheets	10	1.58	Mater. Chem. Front., 2018, 2, 1732-1738.
MoS_2/NiS	10	1.64	Small, 2019, 15, 1803639.
MoS_2/NiS_2 nanosheets	10	1.59	Adv. Sci., 2019, 6, 1900246.
$CoS_x@MoS_2$	10	1.67	Appl. Catal. B Environ., 2019, 247, 107-114.
$NiCo_2S_4$ nanowires	10	1.59	ChemistrySelect, 2019, 4, 1180-1187.
CoS_x/Ni_3S_2	10	1.57	CS Appl. Mater. Interfaces, 2018, 10, 27712–27722.
NiS/NiS_2	10	1.62	J. Mater. Chem. A, 2018, 6, 8233-8237.
e-$(NiCo)S_x/(OH)_x$	10	1.6	Adv. Mater., 2018, 30, 1705538.