

Supplementary Materials

# Bimetallic Cu/Fe Metal-Organic Framework-Based Nanosheet-film via Binder-Free Drop-Casting Route: A Highly Efficient Urea-Electrolysis Catalyst Demonstrating an Ultra-High Current

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**Citation:** Patil, S.A.; Shrestha, N.K.; Inamdar, A.I.; Bathula, C.; Jung, J.; Hussain, S.; Nazir, G.; Kaseem, M.; Im, H.; Kim, H. Bimetallic Cu/Fe Metal-Organic Framework-Based Nanosheet-film via Binder-Free Drop-Casting Route: A Highly Efficient Urea-Electrolysis Catalyst Demonstrating an Ultra-High Current. *Nanomaterials* **2022**, *12*, 1916.  
<https://doi.org/10.3390/nano12111916>

Academic Editor(s): Hanfeng Liang

Received: 27 April 2022

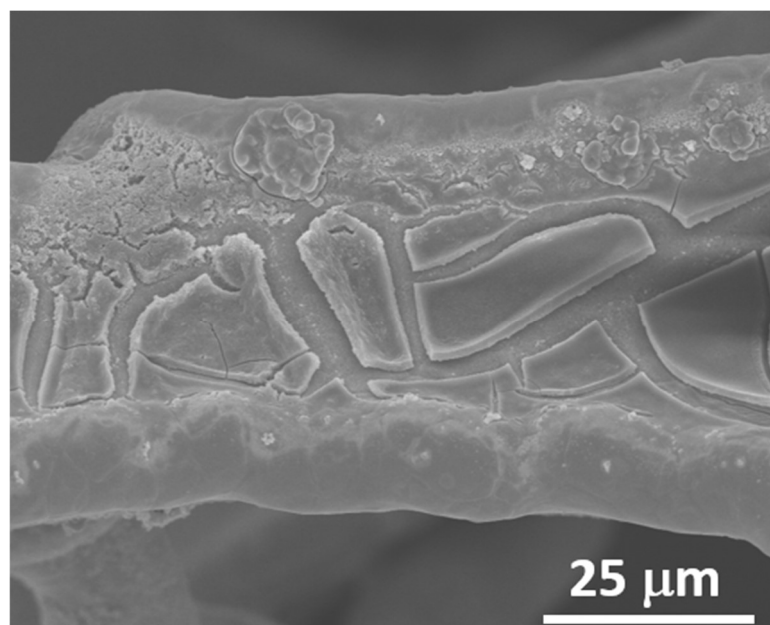
Accepted: 01 June 2022

Published: 03 June 2022

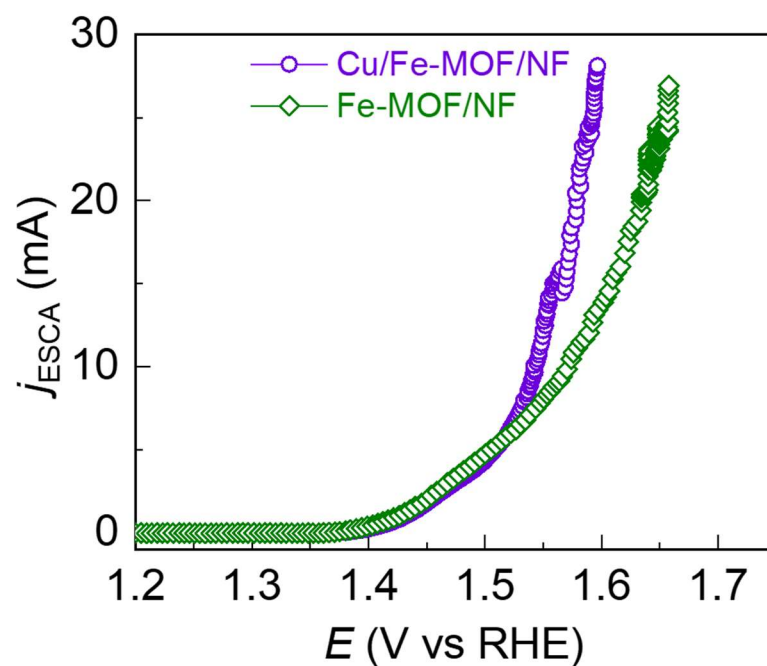
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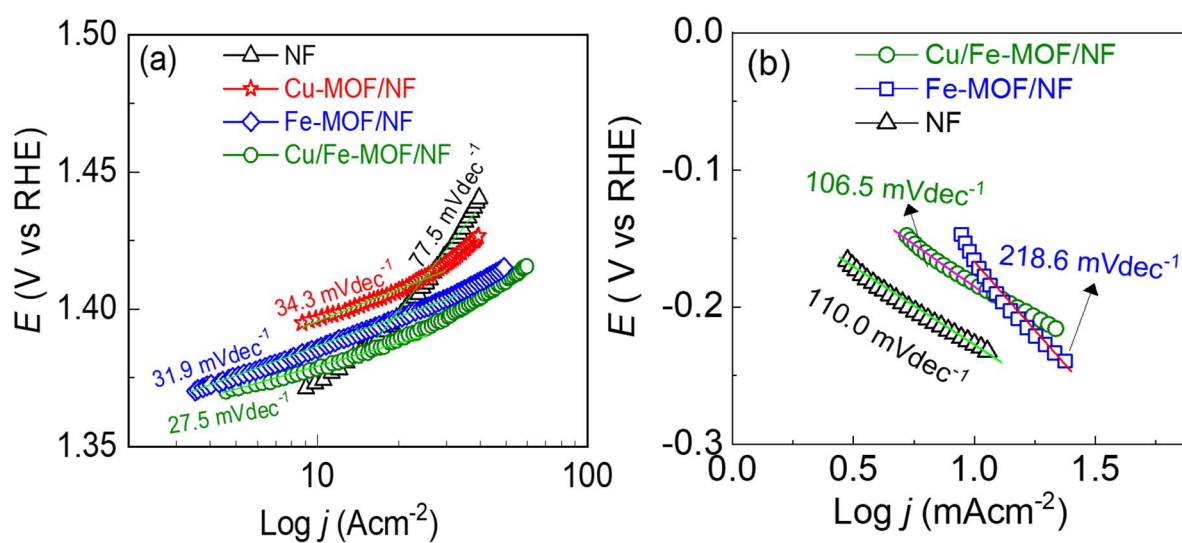
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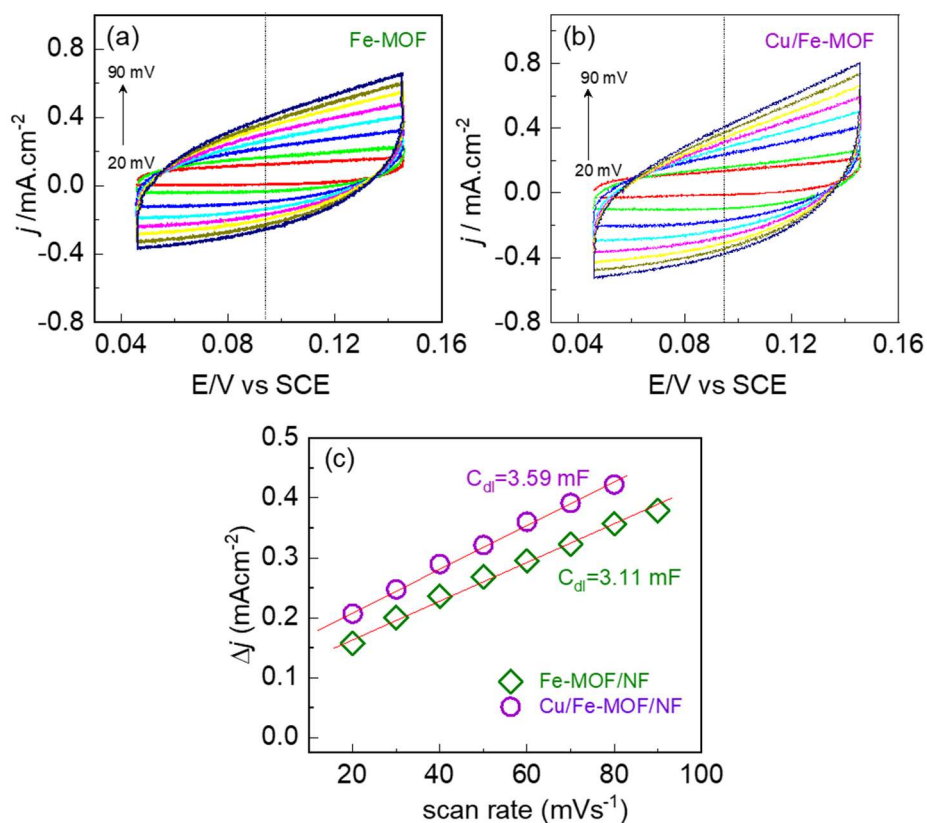
**Figure S1.** SEM image of the drop-casted Cu/Fe-MOF film on a nickel backbone of the nickel foam substrate, showing the sheet-like structure.



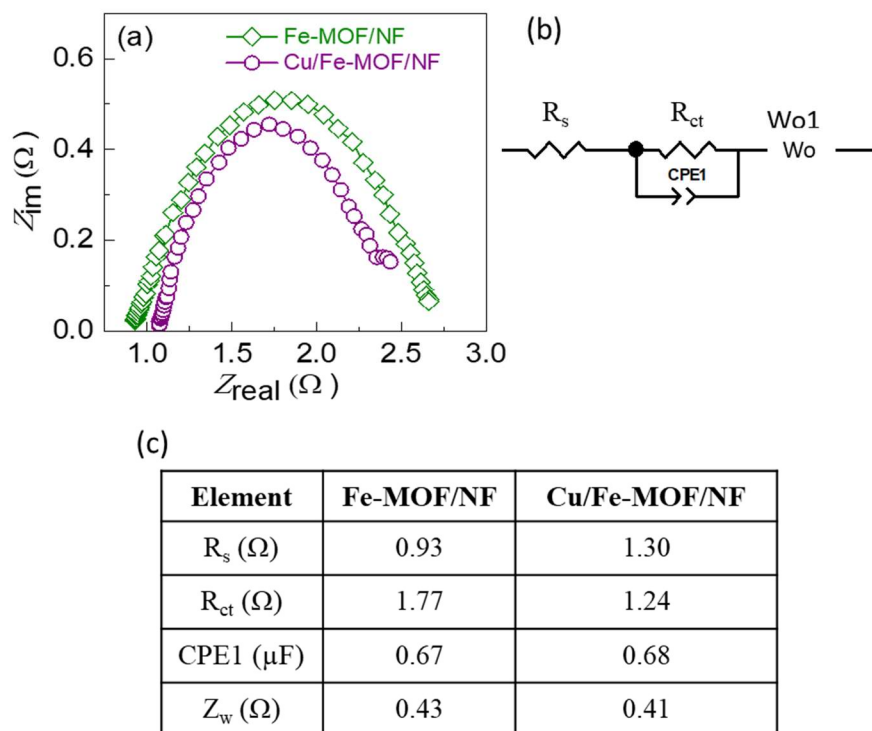
**Figure S2.** ECSA specific LSV polarization curves obtained in 1.0 M KOH containing 0.33 M urea by normalizing the geometrical area-based current density by the ECSA.



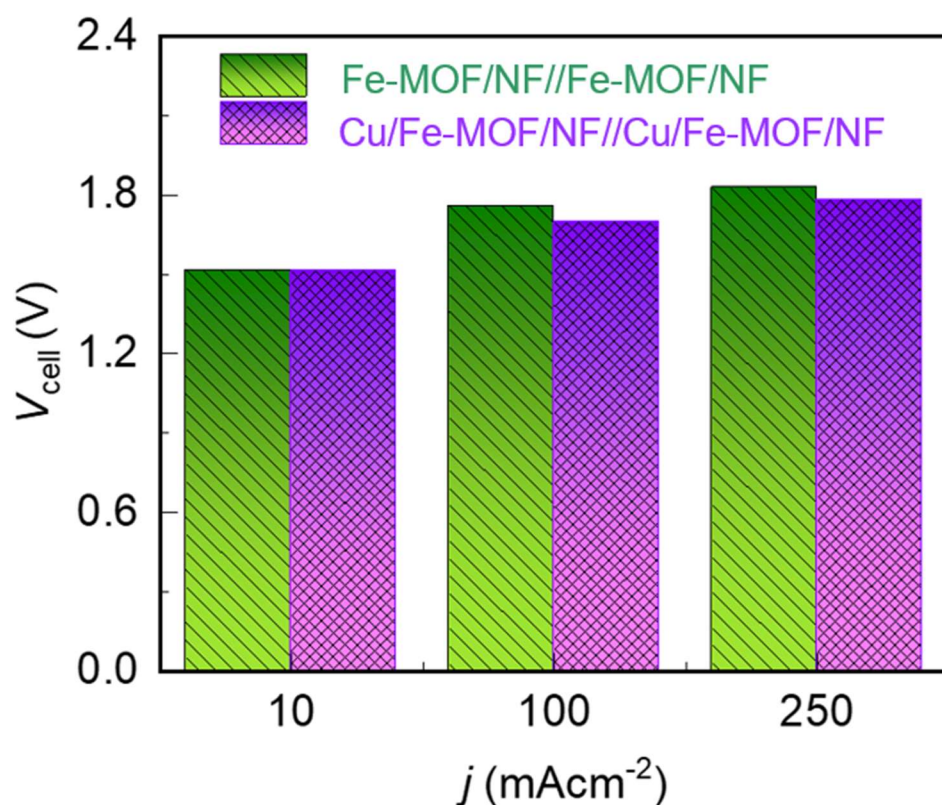
**Figure S3.** (a) Tafel slopes for UOR extracted from the corresponding anodic LSV polarization curves of “Figure 5a”, and (b) Tafel slopes for HER extracted from the corresponding cathodic LSV polarization curves of “Figure 5d”.



**Figure S4.** (a-b) CV plots of the Fe-MOF/NF and Cu/Fe-MOF/NF electrodes, respectively in the non-faradaic region in 1.0 M KOH containing 0.33 M urea at various scan rates ranging from 20 to 90  $\text{V s}^{-1}$ . (c) double-layer capacitance ( $C_{dl}$ ) values estimated from the corresponding CV plots.



**Figure S5.** Electrochemical impedance spectroscopy of the Fe-MOF/NF and Cu/Fe-MOF/NF electrodes. (a) Nyquist plots measured at 1.51 V (vs. RHE), (b) equivalent circuit, and (c) EIS parameter extracted from the Nyquist plots.



**Figure S6.** UOR current density vs  $V_{\text{cell}}$  profile for the overall urea splitting electrolyzers consisting of symmetrical anode and cathode in 1.0 M KOH solution containing 0.33 m urea.

**Table S1.** Comparison of UOR performance for the Cu/Fe-MOF/NF with respect to reported high-performance UOR electrocatalysts.

	Electrode materials	Electrolyte	$j$ (mAcm <sup>-2</sup> )	Cell potential (V) vs RHE	References
	Cu/Fe-MOF/NF	1.0 M KOH + 0.33 M urea	100 500 1000 1500 2500	1.43 1.51 1.54 1.57 1.59	<b>This work</b>
	Fe-MOF/NF	1.0 M KOH + 0.33 M urea	100 500 1000 1500 2500	1.43 1.53 1.59 1.63 NA	<b>This work</b>
1	NiFeRh-LDH	1.0 M KOH + 0.33 M urea	500	1.55	<i>Appl. Catal. B Environ.</i> <b>2021</b> , 284, 119740
2	NiS@Ni <sub>2</sub> S/NiMoO <sub>4</sub>	1.0 M KOH + 0.5 M urea	100 450	1.46 1.78	<i>J. Mater. Chem. A</i> <b>2020</b> , 8, 18055–18063
3	NiFeCo-LDH nanosheets	1.0 M KOH + 0.33 M urea	170	1.55	<i>ACS Sustain. Chem. Eng.</i> <b>2019</b> , 7, 10035-10043
4	NFHC	1.0 M KOH + 0.5 M urea	100	1.40	<i>Sci. Rep.</i> <b>2019</b> , 9, 1–11
5	Ni metal-organic framework	1.0 M KOH + 0.33 M urea	65	1.50	<i>Chem. electrochem</i> <b>2018</b> , 5, 2795-2807
6	V <sub>0</sub> -rich-CoMoO <sub>4</sub> /NF	1.0 M KOH + 0.5 M urea	100	1.51	<i>Appl. Catal. A Gen.</i> <b>2020</b> , 602, 117670.

7	NC-PB@CNT	1.0 M KOH + 0.33 M urea	100	1.41	<i>Chem. Eng. J.</i> <b>2021</b> , 426, 130773
8	NiFe(OH) <sub>2</sub> -SD/NF	1.0 M KOH + 0.33 M urea	100	1.52	<i>J. Colloid Inter. Sci.</i> <b>2019</b> , 557 10-17
9	NiRh/C	1.0 M KOH + 0.33 M urea	183	1.50	<i>Electrochim. Acta</i> <b>2019</b> , 297 715-724
10	Co <sub>3</sub> O <sub>4</sub> @Co <sub>2</sub> P <sub>4</sub> O <sub>12</sub> -300	1.0 M KOH + 0.5 M urea	100	1.60	<i>Int. J. Hydrogen Energy</i> , <b>2019</b> , 44, 24705-24711
11	Ni <sub>2</sub> P/Fe <sub>2</sub> P/NF	1.0 M KOH + 0.5 M urea	100	1.45	<i>J. Colloid Interface Sci.</i> , <b>2019</b> , 541, 279-286.
12	CE-NiFe/NF	1.0 M KOH + 0.33 M urea	100	1.5	<i>Electrochim. Acta</i> , <b>2017</b> , 227, 210-216
13	NiIr-MOF/NF	1.0 M KOH + 0.5 M urea	100 300	1.349 1.350	<i>Chem. Commun.</i> <b>2020</b> , 56, 2151–2154
14	NiMoO-Ar/NF	1.0 M KOH + 0.5 M urea	100 300	1.42 1.52	<i>Energy Environ. Sci.</i> <b>2018</b> , 11, 1890–1897
15	CoS <sub>2</sub> -MoS <sub>2</sub> /NF	1.0 M KOH + 0.5 M urea	100 350	1.33 1.36	<i>Adv. Energy Mater.</i> <b>2018</b> , 8, 1–8.

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