

## Intrinsic metal-support interactions break the activity-stability dilemma in electrocatalysis

Lingxi Zhou<sup>1</sup>, Menghao Yang<sup>2</sup>, Yihong Liu<sup>2</sup>, Feiyu Kang<sup>3,4</sup> and Ruitao Lv<sup>\*1,4</sup>

<sup>1</sup> State Key Laboratory of New Ceramics and Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing, China

<sup>2</sup> Shanghai Key Laboratory for R&D and Application of Metallic Functional Materials, School of Materials Science and Engineering, Tongji University, Shanghai, China

<sup>3</sup> Institute of Materials Research and Shenzhen Geim Graphene Center, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen, China

<sup>4</sup> Key Laboratory of Advanced Materials (MOE), School of Materials Science and Engineering, Tsinghua University, Beijing, China

\*e-mail corresponding: lvruitao@tsinghua.edu.cn

### Significance and Relevance

We report the very first ‘chemical steam deposition (CSD)’ strategy armed with machine learning method to synthesize an integrated ruthenium-titanium-manganese (RuTiMnO<sub>x</sub>) electrode featuring ‘intrinsic metal-support interactions’ (Figure 1). These interactions induced a self-healing mechanism during the OER for H<sub>2</sub> production, thereby enabling both high activity and stability across the full pH range. The CSD strategy enables ‘bottom-up’ atomic-scale nanomaterial synthesis, and ‘intrinsic metal-support interactions’ pave the way to overcome the activity-stability challenge in electrocatalysis.

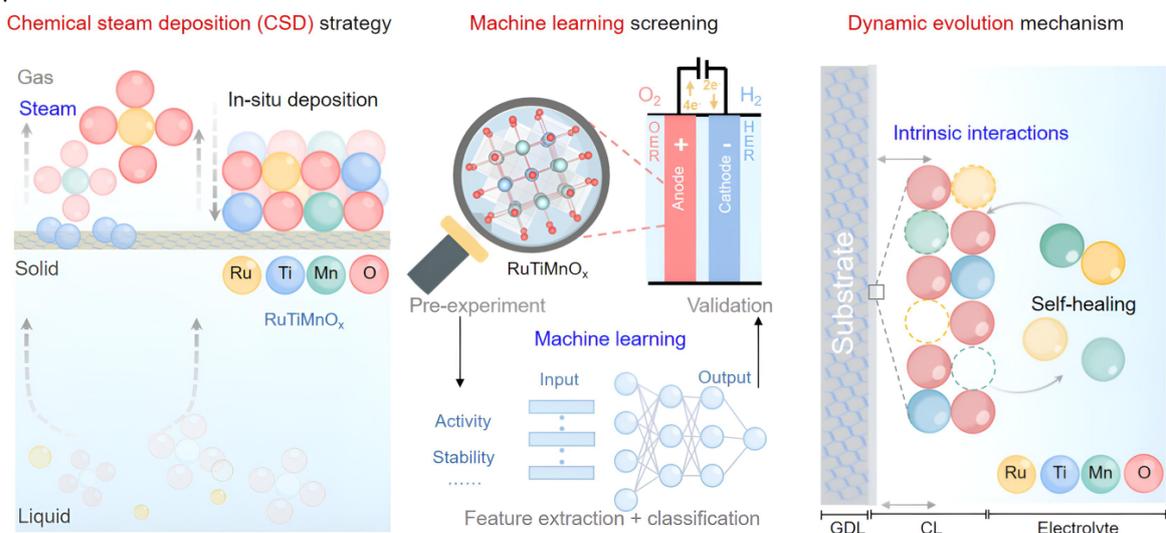
*Preferred and 2<sup>nd</sup> choice for the topic: H<sub>2</sub> storage and transportation, green H<sub>2</sub> production, hydrogen vectors; Advanced process with electrocatalysis and plasma utilization*

*Preferred presentation: Oral preferred or Short Oral*

### Introduction and Motivations

The global energy crisis and climate issues urgently require green energy technologies,<sup>1</sup> with electrocatalysis occupying a pivotal position in clean energy conversion processes essential for future technologies.<sup>2</sup> However, the long-standing dilemma between the activity and the stability of electrocatalysts poses significant constraints on the practical applications, particularly in the case of oxygen evolution reaction (OER) for producing H<sub>2</sub> and other solar fuels.<sup>3</sup>

Here, we report the very first ‘chemical steam deposition (CSD)’ strategy armed with machine learning method to synthesize an integrated RuTiMnO<sub>x</sub> electrode featuring ‘intrinsic metal-support interactions’ (Figure 1). Employing machine learning, we efficiently identified the optimal atomic metal-support ratio considering both activity and stability metrics, avoiding traditional trial-and-error approaches.



**Figure 1.** Chemical steam deposition synthesis, machine-learning-assisted screening and catalysis mechanism study of RuTiMnO<sub>x</sub>.

## Results and Discussion

The resulting electrodes achieved high pH-universal mass activities, 48.5, 112.8 and 74.6 times those of benchmark ruthenium oxides in acidic, neutral and alkaline conditions, respectively; and stable operation for up to 3,000 hours in the challenging pH=0 media, a multi-fold improvement in stability over the reported advanced catalysts (Table 1). Extensive in-situ characterizations combined with theoretical calculations have unveiled the pivotal role of intrinsic metal-support interactions and their dynamic evolution mechanisms in breaking the activity-stability dilemma in OER. Our findings open a new avenue of manipulating intrinsic metal-support interactions to overcome the activity-stability tradeoff dilemma in electrocatalysis.

**Table 1.** Comparison of catalytic performance of different OER catalysts

Media	Sample	$\eta_{10}$ (mV)	Stable Time (h) @ 10 mA cm <sup>-2</sup>	Electrolyte	Data source
Acidic	RuTiMnO <sub>x</sub>	165.2	3000	0.5 M H <sub>2</sub> SO <sub>4</sub>	This work
	RuO <sub>2</sub>	224.1	4	0.5 M H <sub>2</sub> SO <sub>4</sub>	This work
	CoWO <sub>4</sub> -delaminated	288	175	0.5 M H <sub>2</sub> SO <sub>4</sub>	<i>Science</i> 384, 1373 (2024)
	LMCF	370	360	0.1 M HClO <sub>4</sub>	<i>Science</i> 380, 609 (2023)
	Ni-RuO <sub>2</sub>	214	200	0.1 M HClO <sub>4</sub>	<i>Nat. Mater.</i> 22, 100 (2023)
Neutral	RuTiMnO <sub>x</sub>	233.5	1500	0.5 M PBS	This work
	RuO <sub>2</sub>	335.4	5	0.5 M PBS	This work
	Ru-VO <sub>2</sub>	269	60	1.0 M PBS	<i>Adv. Mater.</i> 36, 2310690 (2024)
	Ni-FeWO <sub>4</sub> @WO <sub>3</sub> /NF	235	200	1.0 M PBS	<i>Adv. Mater.</i> 36, 2308925 (2024)
Alkaline	RuTiMnO <sub>x</sub>	185.6	2000	1.0 M KOH	This work
	RuO <sub>2</sub>	239.6	10	1.0 M KOH	This work
	FeCo-N <sub>3</sub> O <sub>3</sub> @C	298	240	0.1 M KOH	<i>Nat. Synth.</i> 3, 878-890 (2024)
	W-718a	320±10	48 h @ 5 mA cm <sup>-2</sup>	0.1 M KOH	<i>Nat. Mater.</i> 23, 252-261 (2024)

## References

1. L. Zhou, Y. Shao, F. Yin, J. Li,\* R. Lv,\* *Nat. Commun.* **2023**, 14, 7644.
2. L. Zhou, Z. Huang, F. Kang, R. Lv,\* *Chem. Eng. J.* **2023**, 458, 141457.
3. L. Zhou, R. Lv\* *J. Energy Chem.* **2022**, 70, 310-331.
4. L. Zhou, M. Guo, Y. Li, et al. *Chem. Commun.* **2019**, 55, 4218.

## Acknowledgements

This work is supported by the National Natural Science Foundation of China (grant numbers 52371228 (R.L.) and 52302302 (M.Y.)), the National Key Research and Development Program of China (grant number 2021YFA1200013 (R.L.)) and Tsinghua University-Toyota Joint Research Center for Hydrogen Energy and Fuel Cell Technology of Vehicles (grant to R.L.). We thank R. X. Zhou and Y. F. Li for technical support, and Q. M. Yuan and G. S. Liao for experimental assistance.