Entropy-Driven Bandgap Engineering in High-Entropy Oxides for Enhanced Solar-Driven Water Evaporation

Yecheng Wang[a], Zeqiong Tan[c], Yang Cao[b], Zubin Wu[a], Shishi Lin[a], Xuehong Jiang[a], Mingxin Zhang[b], Chengcheng Li *[b], Wei Huang *[b].

[a] *National Energy Group Ledong Power Generation Co., Ltd.*

[b] *School of Marine Technology and Equipment, School of Chemistry and Chemical Engineering, Hainan University, Haikou, Hainan 570228, P.R. China.*

[c] *Sansha Marine Center, Ministry of Natural Resources, Haikou, Hainan 570100, P.R. China*

***Corresponding authors**：

Email: licc@hainanu.edu.cn (C. Li)

huangwei@hainanu.edu.cn (W. Huang)

Note S1. Configuration entropy calculation of samples.

We calculated configuration entropy ΔS_{mix} by the following equation:

$$\Delta S_{mix} = -R\left(\sum_{i}^{n} x_i Inx_i\right) \tag{1}$$

Where ΔS_{mix} is the configurational entropy of mixing, R is the gas constant of 8.314 J mol^{-1} K^{-1}, x_i is the molar ratio of component i, and n is the total number of elements involved. The calculated configuration entropy of ZnCdS, ZnCdFeS, ZnCdFeCoS, ZnCdFeCoNiS are 0.69 R, 1.11 R, 1.39 R and 1.61 R, respectively.

Note S2. Layer distance calculation of samples

We calculate the layer spacing by the following equation:

$$2d\sin\theta = n\lambda \tag{2}$$

where d is the spacing between the crystal planes, θ is the angle between the incident X-rays and the corresponding crystal planes, λ is the wavelength of the X-rays, and n is the diffraction series.

Note S3. A detailed, exemplary calculation of the solar-to-vapor energy conversion efficiency for the (FeCoNiCrCu)O sample

$$\eta_{(FeCoNiCrCu)O} = \frac{(1.88\ kg\ m^{-2}\ h^{-1} - 0.28\ kg\ m^{-2}\ h^{-1})(2061\ kJ\ kg^{-1})}{1\ kW\ m^{-2}} \tag{3}$$

$$= \frac{\frac{1.6}{3600} kg\ m^{-2}\ s^{-1} \times 2061\ kW\ s\ kg^{-1}}{1\ kW\ m^{-2}}$$

$$= 91.6\%$$

Table S1. Lattice distortion rate of different samples.

Materials	2θ (XRD)	*d* (Å)	Lattice distortion rate (*ε*, %)
NiO	43.25	2.089	/
(CoNi)O	42.82	2.109	0.96
(FeCoNi)O	42.9	2.105	0.77
(FeCoNiCr)O	43.01	2.1	0.53
(FeCoNiCrCu)O	42.9	2.105	0.77

Table S2. Comparison of the evaporation performance of (FeCoNiCrCu)O with recently reported materials.

Entry	Materials	Evaporation rate (kg m^{-2} h^{-1})	Year	References
1	Mo_2C-C@PAN	1.81	2023	[1]
2	CNTs-CH	1.52	2023	[2]
3	KLC/MF	1.54	2024	[3]
4	PEGDA	1.34	2024	[4]
5	PCPNCs	1.46	2024	[5]
6	JPCF	1.77	2024	[6]
7	CNT10@AAO	1.47	2024	[7]
8	CTS@CTS/CNT	1.55	2024	[8]
9	CBMM	1.81	2024	[9]
10	MoS_2/GO	1.69	2024	[10]
11	PCM	1.85	2024	[11]
12	CF/TiO_2-EPE	1.81	2025	[12]
13	ZPF	1.51	2025	[13]
14	H-ZIF-67@SS	1.62	2025	[14]
15	BTF	1.51	2025	[15]
This work	**(FeCoNiCrCu)O**	**1.88**	**2025**	

References.

1. Li, C.; Cao, Y.; Tian, X.; Shen, Y.; Xiao, J.; Huang, W.; Wang, H.; Liu, Z., Dual-hydrophilic Janus evaporator for Long-term and efficient Bimode solar evaporation. *Chem. Eng. J.* **2023,** *461*, 141954.
2. Wang, X.; Sun, Y.; Zhao, G.-y.; Wang, X.-z.; Qiu, J.-s., Preparation of carbon nanotube/cellulose hydrogel composites and their uses in interfacial solar-powered water evaporation. *New Carbon Mater.* **2023,** *38* (1), 162-172.
3. Li, W.; Li, T.; Deng, B.; Xu, T.; Wang, G.; Hu, W.; Si, C., Fabrication of a facile self-floating lignin-based carbon Janus evaporators for efficient and stable solar desalination. *Adv. Compos. Hybrid Mater.* **2024,** *7* (2), 52.
4. Naghdi, B.; Arabpour Roghabadi, F.; Soleimani-Gorgani, A., Salt-resistant solar water desalination system via surface modification and configuration engineering. *Desalination* **2024,** *577*, 117390.
5. Su, Q.; Yan, J.; Xiao, W.; Liu, Y.; Li, L.; Wang, J.; Huang, X.; Xue, H.; Gao, J., Flexible nanofiber composite solar evaporator for simultaneous interfacial evaporation and heavy metal ion adsorption. *Chem. Eng. J.* **2024,** *490*, 151814.
6. Sun, J.; Xin, Y.; Li, Z.; Sun, B.; Fan, X., Rapid preparation of Janus biomass evaporator by dielectric barrier discharge plasma for high-efficiency desalination and wastewater purification. *Chem. Eng. J.* **2024,** *484*, 149669.
7. Xia, Q.; Pan, Y.; Liu, B.; Zhang, X.; Li, E.; Shen, T.; Li, S.; Xu, N.; Ding, J.; Wang, C.; Vecitis, C. D.; Gao, G., Solar-driven abnormal evaporation of nanoconfined water. *Sci. Adv.* **2024,** *10* (22), eadj3760.
8. Xue, Y.; Han, X.; Xu, D.; Zhang, Z.; Zhu, L.; Wang, S., All-in-one chitosan-based aerogel with a semi-clad structure for solar-driven interfacial evaporation. *Desalination* **2024,** *575*, 117333.
9. Zhang, B.; Chen, H.; Huang, Y.; Liu, Z.; Lau, W.-M.; He, X.; Zhou, D., A coal-based multifunctional membrane for solar-driven seawater desalination and power generation. *Desalination* **2024,** *578*, 117451.
10. Zheng, H.; Fan, J.; Chen, A.; Li, X.; Xie, X.; Liu, Y.; Ding, Z., Enhancing Solar-Driven Water Purification by Multiscale Biomimetic Evaporators Featuring Lamellar

MoS2/GO Heterojunctions. *ACS Nano* **2024,** *18* (4), 3115-3124.

11. Zheng, Z.; Liu, H.; Wang, X., Double-layered hydrogels based on phase change material and pen ink for continuous and efficient solar-driven seawater desalination. *Desalination* **2024,** *574*, 117276.

12. Guo, H.; Zhang, T.; Zhang, X.; Mao, C.; Liu, X.; Wan, S.; Li, Q.; Sun, J.; Dong, S.; Yu, C.; Zhu, Y., Rational design of “ship-in-a-bottle” evaporator with integrated solar evaporation and photocatalytic degradation for sustainable water treatment. *Chem. Eng. J.* **2025,** *506*, 159995.

13. Hu, D.; Zhang, Z.; Zhang, G.; Zhang, G.; Ma, C., Self-Generating Zwitterionic Polyurethane Foam for Solar-Driven Water Evaporation in Complex Environments. *ACS Appl. Mater. Interfaces* **2025,** *17* (11), 17346-17357.

14. Zhang, W.; Liu, J.; Liu, S.; Huang, E.; Ren, S.; Gong, L., Hollow ZIF-67-loaded sandy sediment for enhanced solar-driven water evaporation and PMS-activated pollutant degradation. *Chem. Eng. J.* **2025,** *506*, 159900.

15. Qian, Y.; Cheng, W.; Chen, L., Rapid laser printing of cost-effective, scalable and flexible carbon-based fabrics for solar water evaporation. *Desalination* **2025,** *597*, 118394.