Nano Research

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Highlight

Sub-nanometer high-entropy oxides: Breaking trade-offs in seawater electrolysis

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Cite this article: *Nano Research*, **2026**, *19*, 94908853. https://doi.org/10.26599/NR.2026.94908853

According to the World Energy Council, global hydrogen demand is projected to reach at least 77 million tons per year by 2030 and nearly double to 148 million tons by 2050 [1]. Water electrolysis powered by renewable energy offers a sustainable route to green hydrogen without carbon emissions. Unlike freshwater, which accounts for less than 1% of Earth's water resources, direct seawater electrolysis leverages a nearly unlimited supply and expands the use of marine energy. However, its implementation is hindered by poor catalytic activity and limited lifetime under industrial scale current densities [2]. In detail, vigorous gas evolution at high current densities creates mechanical stress that conventional polymer binders cannot withstand, causing progressive delamination and loss of electrical contact. Meanwhile, partial dissolution of labile elements during prolonged operation triggers structural collapse, destroying the catalytically active architecture. Together, these mechanical and chemical failure modes severely limit catalytic activity and operational lifetime under industrial scale conditions. In a recent work published in *Nature Nanotechnology*, Huang et al. [3] demonstrated a 14-element high-entropy oxide sub-nanowire that defies conventional trade-offs. By operating at the sub-1 nm scale, their catalyst achieves low-temperature synthesis, intrinsic self-adhesion, and exceptional activity with long-term stability, demonstrating great promise for binder-free industrial seawater electrolysis. This is not merely another high-entropy catalyst but a rethinking of how nanoscale design can reconcile competing properties.

1. Low-temperature synthesis. The synthesis of high-entropy oxides (HEOs) has traditionally been a high-temperature art, typically requiring calcination above 1000 K to overcome the enthalpic penalty of mixing multiple elements [4]. In striking contrast, this work achieves a single-phase, 14-element high-entropy oxide sub-nanowire (14-HEO SNW) under mild hydrothermal conditions at just 160 °C (Fig. 1(a)). Previous reports of low-temperature HEO formation have often attributed the result to kinetic trapping or ligand effects [5, 6]. This extraordinary deviation from conventional thermodynamics is driven by a dimensionality-induced thermodynamic reconfiguration. As the wire diameter is compressed to the sub-nanometer limit of ~ 1.2 nm (Fig. 1(b)), the proportion of surface atoms escalates dramatically. These under-coordinated surface atoms exhibit softened phonon modes, generating a substantial surface entropy contribution. As described by the Gibbs free energy equation $\Delta G = \Delta H_{mix} - T(\Delta S_{configuration} + \Delta S_{surface})$, the $\Delta S_{surface}$ term becomes comparable to, and even dominant over, the configurational entropy ($\Delta S_{configuration}$) at this scale [7]. This surface-entropy-dominated regime drastically lowers the free energy barrier for single-phase formation. Combined with confined one-dimensional growth, this mechanism enables the low-temperature synthesis of 14-HEO SNWs. Rather than being a mere source of defects, the surface entropy arising from sub-nanostructures serves as a thermodynamic engine that stabilizes multi-component systems, enabling a new low-temperature synthesis paradigm.

2. Intrinsic self-adhesion. The development of adhesive sub-nanometer materials began with the discovery that ultrathin inorganic nanowires (diameters < 1 nm) exhibit polymer-like flexibility and self-assembly. Early work on rare earth hydroxide nanostructures (e.g., GdOOH) revealed ligand-driven gelation and conformational diversity [8]. Subsequently, Ca-POM (calcium polyoxometalate) sub-nanometer wires were developed as versatile adhesives [9]. The atomic-molecular origins of their adhesion features with high strength, reversibility, and environmental stability are proposed to arise from ultrahigh flexibility and multilevel interactions, including electrostatic forces between Ca^{2+} and $PW_{12}O_{40}^{3-}$ clusters, van der Waals forces from organic ligands such as oleylamine, and entanglements among nanowires. Breaking the current limitation of "pure adhesion" and developing functional adhesive systems will truly drive sub-nanometer materials from "structural novelty" toward practical applications. Unlike conventional electrocatalysts that require polymer binders like Nafion, which increase resistance and degrade under high current densities, 14 HEO SNWs form an entangled network in solvent that exhibits polymer-like rheology. This adhesion strength remains stable after prolonged immersion in aqueous solutions across pH 1 to 14 and in seawater (Fig. 1(c)). Two stainless steel blocks bonded solely by these nanowires can lift a 10 kg weight, achieving an adhesion strength of 0.84 MPa on steel, approximately 982 times higher than that of Nafion (Fig. 1(d)). Strong interfacial adhesion arises from the high entropy distortion that turns the inorganic sub-nanowires into an intrinsic adhesive. Incorporating 14 metal elements with varying ionic radii and coordination preferences generates extensive local strain and abundant unsaturated metal–oxygen dangling bonds densely distributed across the sub-

Received: May 14, 2026
Accepted: May 18, 2026

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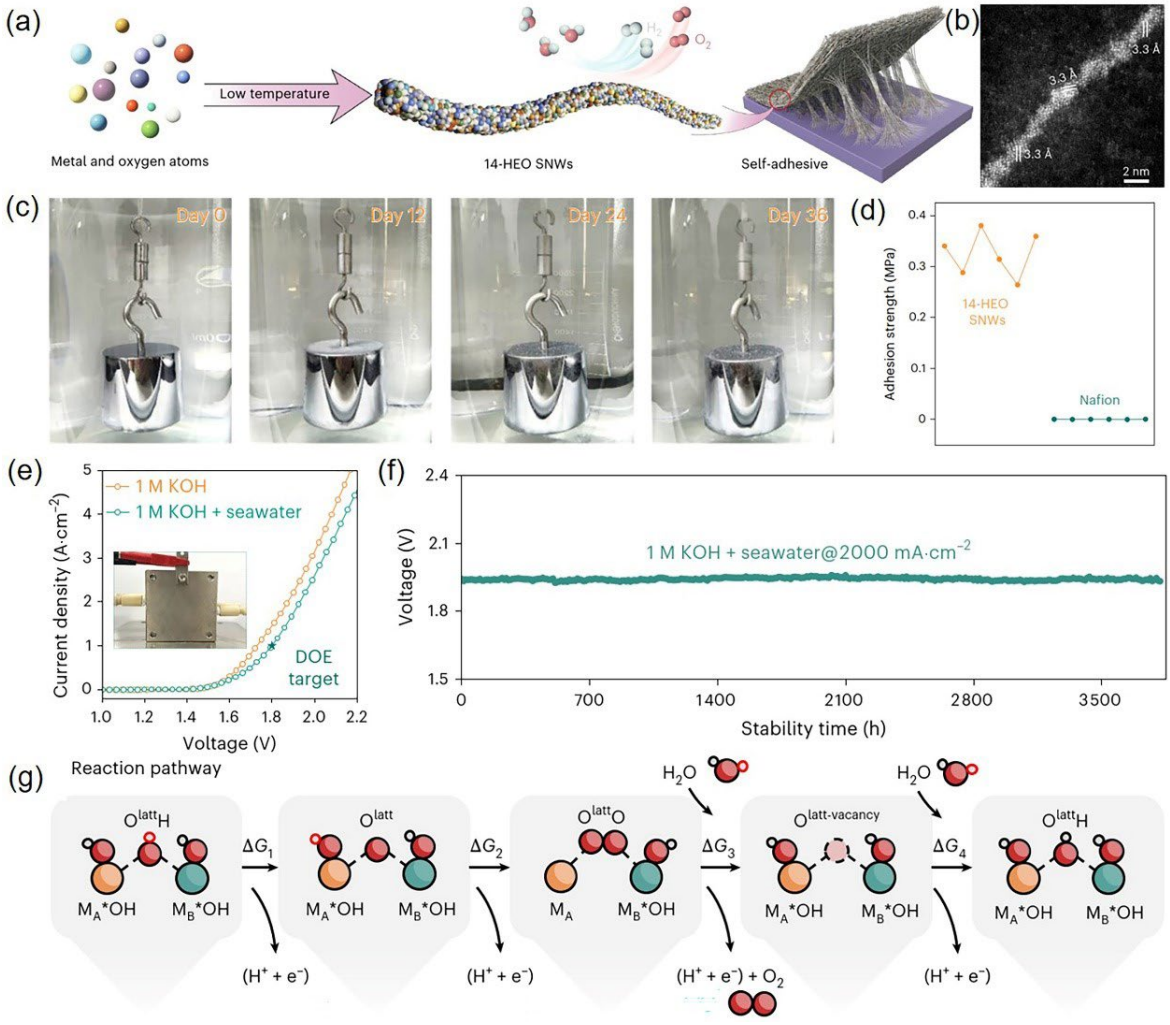

Figure 1 (a) Schematic illustration of the synthesis pathway of self-adhesive 14-HEO SNW. (b) Aberration-corrected high-angle annular dark field scanning TEM (HAADF-STEM) image. (c) Adhesion behavior under macroscopic testing conditions in KOH + seawater. (d) Comparative assessment of adhesion performance using commercial Nafion as reference. (e) Room-temperature linear sweep voltammetry (LSV) curves of for 14-HEO SNWs||Pt/C electrolyser. (f) Long-term durability of electrolyser operated at 2000 mA·cm^{-2} in 1 M KOH + seawater. (g) Reaction pathways for LOM on M_A–O–M_B (Cu–O–Cr). Reprinted with permission from Ref. [3], © Huang, Y. et al., under exclusive licence to Springer Nature Limited 2026.

nanowire surface. These metal–oxygen bonds function similarly to polar groups in polymer adhesives, yet the fully inorganic framework offers superior chemical stability and mechanical strength, transforming structural imperfections into a functional advantage for robust catalyst–electrode interfaces.

3. High activity and stability. Once the immobilization issue is resolved, the intrinsic oxygen evolution reaction (OER) activity of the material becomes the next critical test. The 14 HEO SNWs achieve an overpotential of only 129 mV at 10 mA·cm^{-2} in 1 M KOH and operate stably for 4700 h at 1000 mA·cm^{-2}. In alkaline seawater, the overpotential at 1000 mA·cm^{-2} is 306 mV, well below the 480 mV threshold for chlorine evolution, with stable operation for 4400 h. Integrated into an anion exchange membrane electrolyzer, the system delivers 1000 mA·cm^{-2} at 1.80 V at room temperature (Fig. 1(e)) and 3000 mA·cm^{-2} at 1.70 V and 80 °C in 1 M KOH with seawater. The continuous operation can stabilize at 2000 mA·cm^{-2} for 3819 h under ambient conditions (Fig. 1(f)). The hydrogen cost reaches as low as 0.82 USD substantially lower than the 2026 target of 2.00 USD set by the U.S. Department of Energy (DOE). Mechanistically, *in situ* Raman and isotope labeled differential electrochemical mass spectrometry (DEMS) confirm a lattice oxygen mechanism (LOM). Density functional theory (DFT) identifies the Cu–O–Cr dual metal center as the optimal active site. Cu stabilizes lattice oxygen while Cr activates it, and their complementary synergy within the high entropy framework achieves superior catalytic performance (Fig. 1(g)). In addition, the HEO host lattice is also critically important. The high entropy framework allows copper to adopt a favorable four-coordination planar geometry, whereas the crystalline CoOOH lattice forces it into a less active octahedral coordination. Additionally, selective dissolution of Mo, Cd, Mg, and Al during OER leaves the nanowire morphology intact, and the remaining ten elements reconstruct into a stable high entropy oxyhydroxide. This dynamic adaptation is key to the long-term stability of high entropy systems under operating conditions.

Taken together, by integrating low-temperature synthesis, intrinsic self-adhesion, and exceptional catalytic stability within a single 14-element oxide sub-nanowire, this work breaks the traditional trade-offs that have long plagued high-entropy materials and seawater electrolysis. The surface-entropy driven synthesis

redefines how metastable phases can be stabilized, while the high-entropy architecture turns structural distortion into a functional advantage for both adhesion and catalysis. This is not merely a new catalyst, but a design paradigm that links thermodynamics, mechanics, and electrochemistry at the sub-nanometer scale.

Acknowledgements

This work is supported by the National Natural Science Foundation of China (No. 92477134) and Provincial Natural Science Foundation of Shandong (No. ZR2023ME014).

Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

H. Q. W.: Project administration, funding acquisition, writing manuscript. The author has approved the final manuscript.

Use of AI statement

During the preparation of this work, the authors used DeepSeek in order to prevent low-level errors in grammar and spelling. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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