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Harnessing a transient liquid-metal flux for the ultrafast photoforging of compositionally complex oxide nanoarchitectures

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ABSTRACT

Synthesizing compositionally complex oxides (CCOs) from thermodynamically immiscible elements is fundamentally constrained by phase segregation during conventional prolonged thermal processing, which also degrades the underlying substrate. Here, we report an ultrafast synthesis strategy, termed photoforging, that couples a transient eutectic gallium–indium liquid-metal flux with a single 20 ms intense pulsed light (IPL) treatment. Under IPL, the flux ruptures and wets the nickel fiber surfaces, uniformly delivering and homogenizing disparate transition-metal precursors while forming a flash-induced Ni–Ga intermetallic layer that monolithically anchors the oxide coating; the concurrent rapid heating and quenching kinetically trap up to ten cations within a metastable, single-phase NiO rock-salt lattice. This process yields CCO nanoarchitectures—including vertically oriented, ultrathin nanosheets in select compositions—directly integrated onto the substrate. Exploiting this compositional versatility, we custom-design a binder-free high-surface-area quinary oxide electrode for water splitting that delivers exceptional activity and durability for both hydrogen and oxygen evolution reactions—one of many reaction-specific combinations accessible through this platform.

Multicomponent transition metal oxides, particularly compositionally complex oxides (CCOs), have emerged as a transformative class of functional materials that offer a vast design space to tailor synergistic elemental interactions.^{1, 2} These materials are particularly promising for advanced catalysis³⁻⁶ and energy storage,^{7, 8} where a multi-cationic lattice can provide diverse adsorption environments with optimized binding energies.^{9, 10} In high-entropy oxides (HEOs), atomic-scale homogenization is primarily achieved through entropy-driven thermodynamic stabilization, which strictly constrains them to near-equimolar compositions of 5 or more elements.¹¹⁻¹³ However, unlocking the most optimal catalytic configurations for specific target reactions inherently demands the freedom to tune non-equimolar elemental ratios and accommodate diverse crystal symmetries, thereby precisely and finely controlling local lattice strain and electronic states.^{3, 14, 15}

Venturing into this broader CCO space, however, means forfeiting the robust entropy-driven stabilization of HEOs, rendering the synthesis exceptionally challenging.^{16, 17} Without such a driving force, integrating diverse elements – which typically exhibit severe mismatches in valence states, coordination preferences, and ionic radii – triggers a strong tendency toward phase segregation.^{18, 19} While conventional synthesis relies on prolonged high-temperature treatments to overcome this substantial mixing enthalpy,¹⁶ such equilibrium-driven conditions inevitably provoke structural coarsening and the severe loss of high-surface-area active sites.^{20, 21} Rapid thermal processing^{20, 22} and liquid-metal-assisted synthesis^{23, 24} have separately enabled kinetic trapping and multielement mixing, yet combining these functions with anisotropic oxide growth and direct substrate integration remains underexplored.

Herein, to bypass these thermodynamic bottlenecks, we developed a millisecond photoforging strategy which effectively decouples atomic-scale homogenization from structural coarsening by

combining a liquid-metal (LM) flux with intense pulsed light (IPL) processing. In this system, eutectic gallium-indium (EGaIn) acts as a transient solvent to drastically lower kinetic barriers for multi-elemental mixing,^{23, 25-27} while IPL-driven flash-thermal shock kinetically traps the cationic mixture into a nonequilibrium homogeneous phase within milliseconds.^{22, 28, 29} Concurrently, rapid IPL-activated wetting and interfacial alloying between the LM flux and the underlying Ni fiber induce substrate-participatory cation diffusion. This process spontaneously yields vertically oriented, ultrathin 2D nanostructures monolithically anchored to a thin Ni–Ga intermetallic layer on the Ni substrate. We designate this unique class of materials as structured compositionally complex oxides (SCCOs). For quinary FeCoNiCuZn SCCOs, microscopy, electron diffraction, and Raman spectroscopy support a NiO-derived rock-salt local structure with nanoscale cation co-localization. A systematic screen of all unary-to-quinary combinations in the Fe–Co–Ni–Cu–Zn set maps both nanosheet-forming and non-forming compositions, and the process also yields a denary nanosheet formulation containing ten co-localized metal species. As a proof-of-concept, the binder-free quinary SCCO electrodes exhibit exceptional activity and durability during both half-cell hydrogen and oxygen evolution reactions (HER and OER) in alkaline media. By coupling transient-flux mixing with rapid heating, quenching, and substrate participation, this platform provides a route toward systematic compositional exploration and the direct synthesis of multimetallic oxide electrodes, which can be further customized for specific target reactions.

Results and discussion

The synthetic procedure for SCCO is depicted in **Figure 1a**. First, using a tip sonicator, transition-metal-alloyed Ga₂O₃-capped EGaIn microparticles (LM@TM) were generated in an ethanolic solution containing various transition metal chlorides, driven by galvanic exchange and

passive oxide formation.³⁰ A piece of Ni fiber paper (NFP) was dip-coated in this LM@TM suspension and subjected to IPL photoforging, which delivers ultrafast thermal transients (Fig. 1b, heating: 5×10^4 K/s; cooling: 10^4 K/s). While conventional furnace heating produces bulky, faceted oxide grains (Fig. 1c and Supplementary Fig. 1), the LM-IPL process instantaneously sculpts vertically oriented, 2D nanosheets (Fig. 1d and Supplementary Fig. 2). High-resolution transmission electron microscopy (HR-TEM) and selected area electron diffraction (SAED) of a SCCO flake scraped from NFP in Figure 1e confirm ultrathin sheets of face-centered cubic (FCC) NiO lattices. Crucially, high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) and energy-dispersive X-ray spectroscopy (EDS) mapping prove a homogeneous distribution of Fe, Co, Ni, Cu, Zn, and O within the nanosheets, whereas Ga localizes at the substrate interface (Fig. 1f and Supplementary Figs. 3 and 4). This spatial separation proves that Ga acts exclusively as a transient liquid flux to mediate atomic mixing and substrate wetting, without contaminating the final SCCO flake, yielding a chemically homogeneous multi-cationic framework. The oxidation states of each element and NiO lattices were further confirmed by X-ray photoelectron spectroscopy (XPS) and Raman analyses (Supplementary Figs. 5 and 6). Taken together, these observations corroborate that LM-IPL successfully circumvents the inevitable limitations of existing high-temperature methods for CCO and HEO generation (Supplementary Table 3).

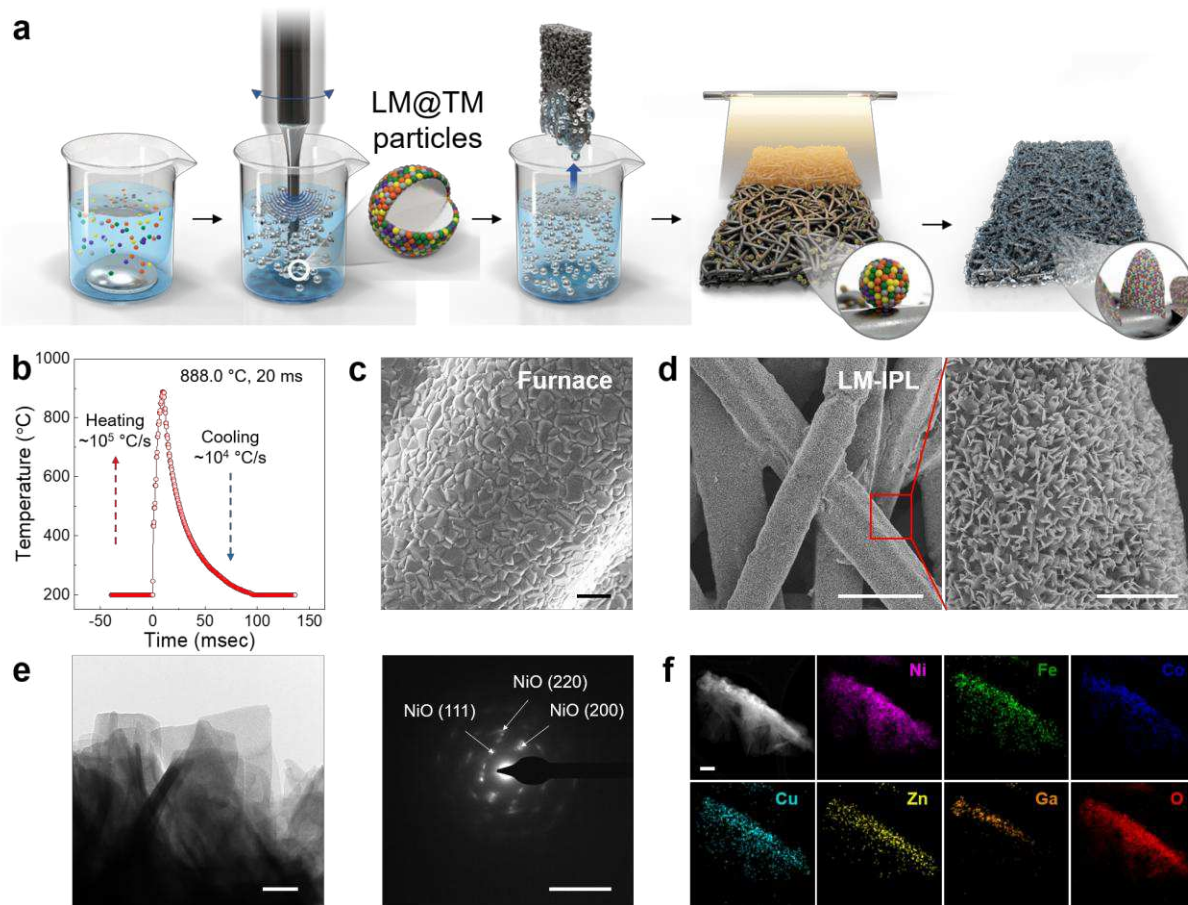


Fig. 1 | Synthesis and characterizations of SCCO. **a**, Schematic illustration of the LM-IPL process. **b**, Temperature-time (T-t) profile under 400 V, 20 ms, and 1 shot at $t=0$. **c**, SEM image of LM@TM coated on Ni fiber paper sintered in a furnace at 900 °C for 1 h. Scale bar: 3 μ m. **d**, SEM image of LM@TM-coated Ni fiber paper photoforged with IPL process. Scale bars: 30 μ m (left) and 3 μ m (right). **e**, HR-TEM image and corresponding SAED pattern of SCCO. Scale bars: 100 nm (left) and 10 nm^{-1} (right). **f**, HAADF-STEM image with EDS elemental maps demonstrating the uniform distribution of Ni, Fe, Co, Cu, Zn, Ga, and O. Scale bar: 100 nm.

A series of control samples were examined by SEM to elucidate the unique flash-flux synergy governing SCCO growth. Before IPL, the precursor consisted of LM@TM particles along with TM chloride residues (**Fig. 2a**). When IPL was applied to the salt residues in the absence of the LM flux, only a smooth coating on the Ni fiber was observed, exhibiting compositional zoning as alternating contrasts (**Fig. 2b**) and EDS maps (**Supplementary Fig. 7**). Conversely, applying IPL

to LM@TM particles on a non-metallic carbon fiber paper resulted in the evolution of spherical alloys with homogeneous compositions (Fig. 2c and Supplementary Fig 8). Together, these specific morphological deviations prove that the hierarchical SCCO architecture is basically driven by substrate-participatory structuring, critically dependent on the Ni–Ga interfacial amalgamation.

The SCCO morphology is also highly sensitive to IPL conditions (Fig. 2d-g). If the IPL power is too weak (300 V and 18.8 J cm^{-2}), the thermal input is insufficient for the entire LM@TM-coated NFP surface to reach the amalgamation temperature and to activate the diffusive environment, resulting in localized SCCO formation only over limited areas (Fig. 2d). Conversely, an excessive peak temperature (450 V, 50.0 J cm^{-2}) severely exacerbates the transient dissolution at the Ni–Ga interface, provoking marked coarsening and partial framework collapse (Fig. 2f). This behavior is consistent with the Ni-Ga binary phase relation, in which interfacial alloying lowers the local melting point and accelerates sintering at excessive temperatures.³¹ Alternatively, increasing the pulse count to three elevates the third peak temperature to $\sim 1000 \text{ }^{\circ}\text{C}$ and prolongs thermal exposure, which drives unwanted secondary grain growth and severely degrades the effective surface area (Fig. 2g). Together, these results show that the precise tuning of IPL thermal transients and the coupled synergy of the LM flux and the metallic substrate are critical for forming high-surface-area SCCO nanostructures.

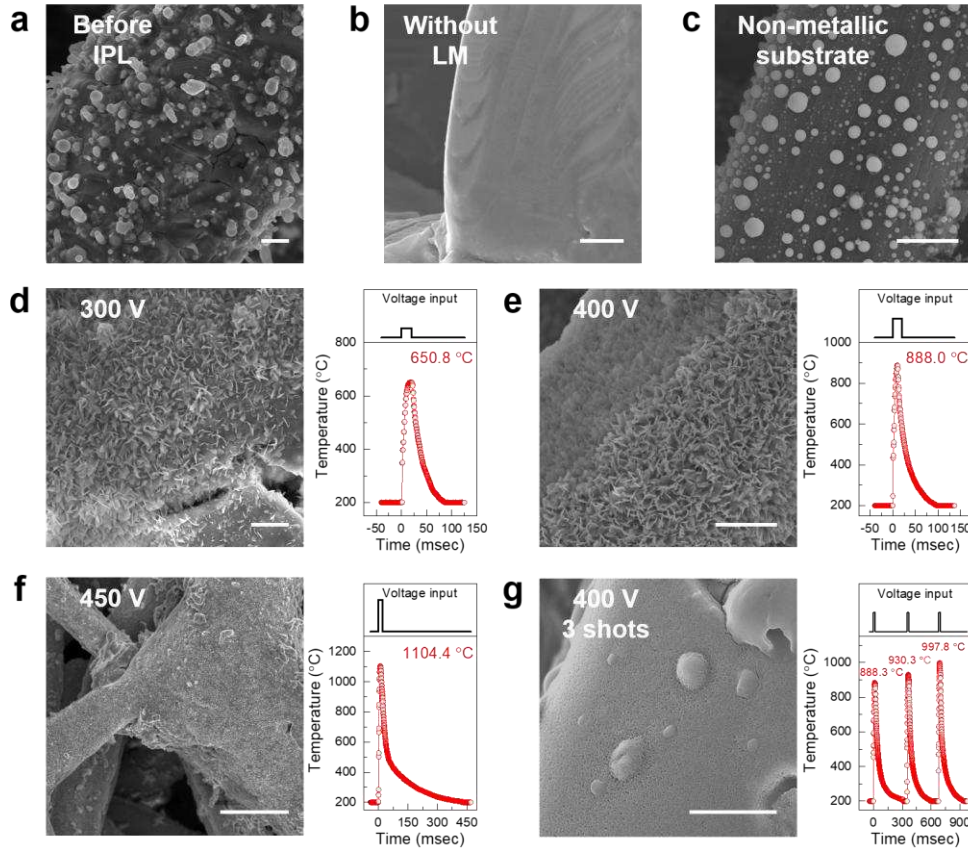


Fig. 2 | Mechanistic investigation and morphology control of SCCO. SEM images **a**, LM@TM-coated Ni fiber before IPL treatment, **b**, Ni fiber paper coated with TM chlorides without LM carriers, after IPL, and **c**, LM@TM-coated carbon fiber paper after IPL treatment. Scale bars: 2 μm . **d–g**, SEM images and corresponding T-t profiles showing morphology evolution on LM@TM-coated Ni fiber paper after various IPL conditions: **(d)** 300 V and 20 ms pulse, **(e)** 400 V and 20 ms pulse, **(f)** 450 V and 20 ms, and **(g)** three pulses of 400 V and 20 ms. Scale bars: 2 μm (d, e, g) and 20 μm (f).

Based on these insights, we propose a three-step photoforging mechanism (Fig. 3). First, the IPL flash melts the LM@TM precursors into a transient Ga flux which instantaneously wets the Ni substrate, inducing interfacial amalgamation. Second, this confined liquid zone enables rapid cation diffusion and homogenization. Driven by extreme thermal gradients, nucleation completely bypasses equilibrium, strongly favoring anisotropic 2D growth over isotropic coarsening. HR-TEM and FFT analyses confirm this non-equilibrium pathway, revealing growth along the [011]

zone axis and the robust retention of high-surface-energy $\{110\}$ facets—structures typically disfavored under near-equilibrium conditions (Supplementary Fig. 9). Finally, ultrafast cooling (10^4 K/s) kinetically traps this metastable morphology, forcefully preventing structural relaxation into flatter, thermodynamically favored aggregates. Ultimately, the LM flux serves as the essential transient solvent, while the IPL flash acts as the kinetic trigger sculpting the hierarchical SCCO architecture.

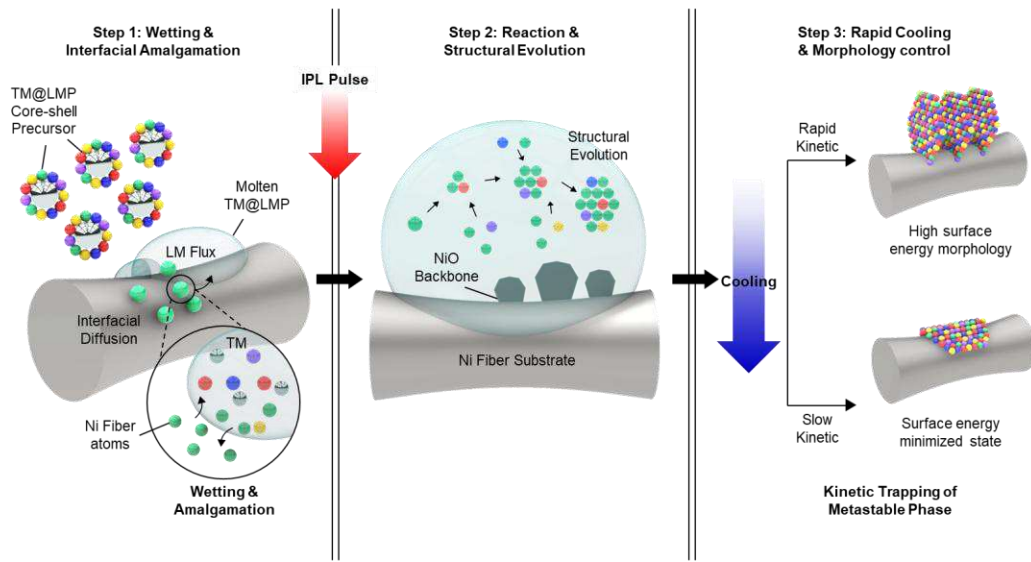


Fig. 3 | Proposed LM-IPL photoforging mechanism for SCCO formation via interfacial Ni–Ga alloying, flux-mediated multication homogenization and 2D oxide growth, and rapid-quench kinetic trapping.

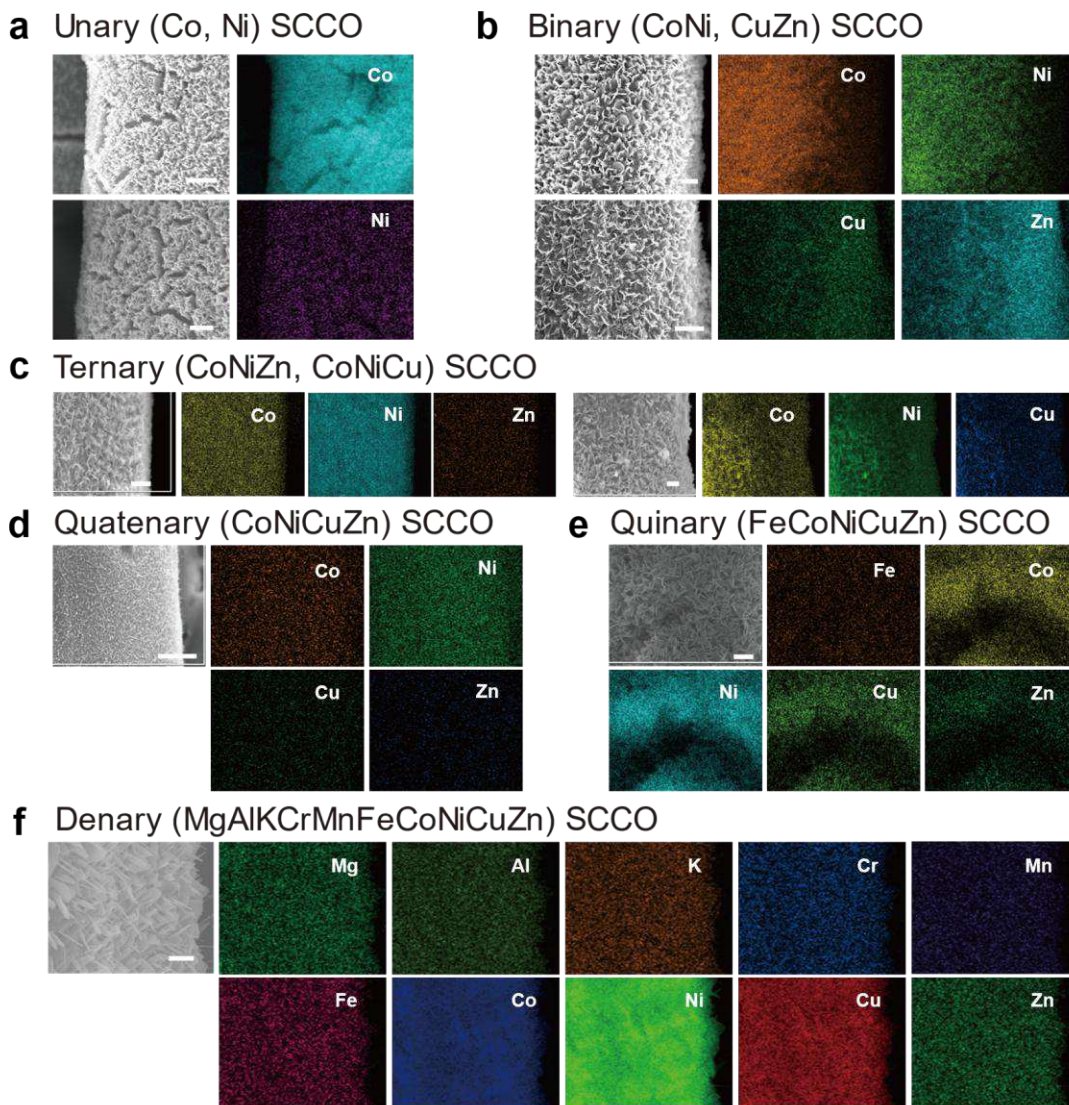


Fig. 4 | Compositional versatility of the LM-IPL platform. SEM images and corresponding EDS elemental maps of multi-metallic oxides synthesized by LM-IPL, ranging from unary to denary (10-element) compositions. Scale bars: 1 μm .

Demonstrating the exceptional compositional versatility of this platform, we synthesized a high-throughput library ranging from unary to denary (10-element) multimetallic oxides (Fig. 4). While the majority of combinations readily yielded the desired hierarchical SCCO architecture along with compositional homogeneity, a notable phase-stabilization threshold emerged in Fe-containing systems (Supplementary Table 4 and Supplementary Fig. 10). In low-entropy regimes

(unary to quaternary), although the constituent elements mixed thoroughly, the limited solubility of Fe in the NiO rock-salt lattice triggered local phase segregation into an inverse spinel (e.g., NiFe_2O_4), which hindered the anisotropic 2D nanosheet growth.³² However, as configurational complexity expanded to five or more cations, the hierarchical 2D morphology forcefully re-emerged despite the presence of Fe. This confirms that high compositional complexity effectively stabilizes the metastable homogeneous framework against thermodynamic segregation. Pushing this limit, we successfully forged a highly uniform denary SCCO containing ten distinct cations. Crucially, the ultrafast synthesis and rapid experimental turnaround of the LM-IPL process establish this platform as an exceptionally powerful tool for high-throughput screening of vast compositional spaces and unlocking novel multimetallic interactions.

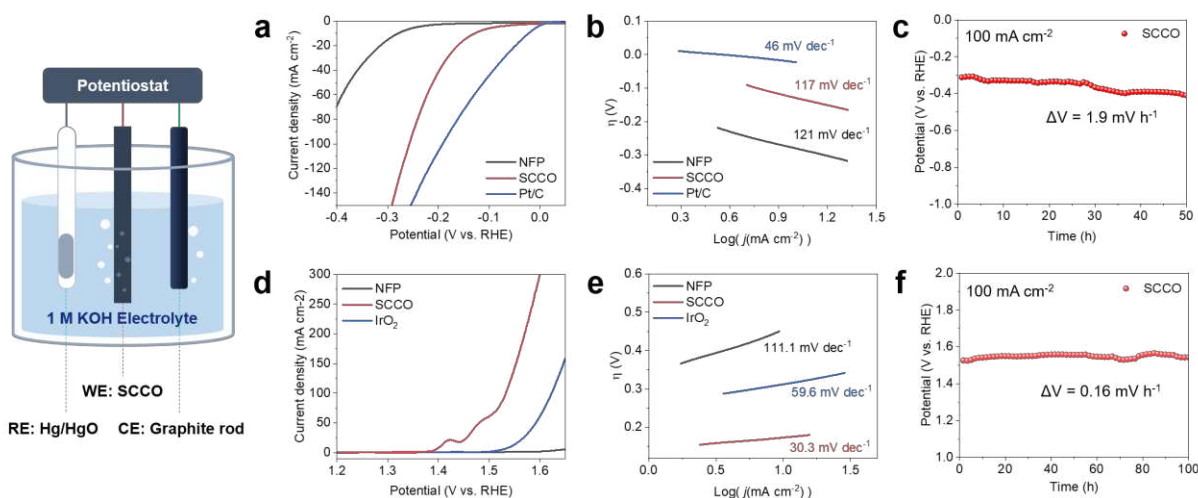


Fig. 5 | Electrocatalytic performance for bifunctional water splitting. **a**, HER polarization curves of SCCO in 1.0 M KOH, compared with Pt/C and bare Ni fiber paper. **b**, Corresponding HER Tafel plots. **c**, HER durability at a constant current density of -100 mA cm^{-2} . **d**, OER polarization curves of SCCO in 1.0 M KOH, compared with IrO_2 and bare Ni fiber paper. **e**, Corresponding OER Tafel plots. **f**, OER durability at a constant current density of 100 mA cm^{-2} .

Harnessing this structural and compositional complexity, we evaluated a quinary SCCO (FeCoNiCuZn oxide) as a bifunctional electrocatalyst for alkaline water splitting.³³⁻³⁵ The

monolithic, binder-free SCCO@NFP electrode exhibits outstanding HER and OER activities, dramatically outperforming bare Ni substrates and rivaling or exceeding commercial benchmarks. For HER, it approaches the activity of Pt/C with a Tafel slope of 117 mV dec⁻¹ and negligible degradation over 50 h at -100 mA cm⁻² (Fig. 5a-c); the instability observed in Figure 5c is primarily attributed to the degradation of the graphite counter electrode, rather than the SCCO catalyst (Supplementary Fig. 11). The OER performance is even more exceptional, surpassing commercial IrO₂ with a significantly lower Tafel slope (30.3 vs. 59.6 mV dec⁻¹, Fig. 5d and e), reflecting substantially accelerated OER kinetics. Remarkably, the catalyst sustains 100 mA cm⁻² for 100 h with a virtually imperceptible voltage decay of 0.16 mV h⁻¹ (Fig. 5f). This superb bifunctionality and extreme durability stem directly from the combination of a maximized electrochemically active surface area (~3.5 times higher, Supplementary Fig. 12) and the suppressed cation mobility inherent to the highly strained, compositionally complex rock-salt lattice, effectively resisting degradation under prolonged alkaline operation.³⁶

Conclusion

In summary, photoforging establishes a general strategy for overcoming the thermodynamic limits that have long constrained compositionally complex oxide synthesis, using a transient liquid-metal flux to kinetically trap immiscible elements into unified SCCO architectures. This emerges from the deliberate synergy of three factors: the intrinsic Ga–Ni wetting affinity to uniformly anchor CCO coating to the substrate, the liquid metal's capacity to homogeneously carry disparate cations, and a thermal cycle confined to tens of milliseconds—each indispensable, together enabling substrate-directed growth that would otherwise be virtually impossible. More broadly, by turning immiscibility into a tunable design axis rather than a constraint, photoforging possibly compresses the search for multimetallic materials from a slow, trial-and-error process into

a millisecond-per-sample experiment—a capability we demonstrate here for water splitting, but equally suited to screening compositions for batteries, sensors, and catalysis at large.

Methods

Materials

Zinc chloride (ZnCl_2 , $\geq 98\%$), iron(III) chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\geq 98\%$), cobalt(II) chloride hexahydrate ($\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, $\geq 98\%$), nickel(II) chloride hexahydrate ($\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$, $\geq 98\%$) and copper(II) chloride dihydrate ($\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, $\geq 98\%$) were purchased from Sigma-Aldrich, eutectic gallium-indium (EGaIn, 99.99%) was purchased from Changsha Rich Nonferrous Metals Co., Nickel fiber paper and carbon fiber paper were purchased from Dioxide Materials, ethanol ($\text{C}_2\text{H}_5\text{OH}$, 99.5%) and potassium hydroxide (KOH, 95%) were purchased from Daejung Chemicals & Metals Co. Commercial Pt/C (46.5 wt%, TANAKA Chemical Corporation) and IrO_2 (99.99%, Alfa Aesar) were used as benchmark electrocatalysts. All aqueous solutions and electrolytes were prepared using ultrapure water (18.2 $\text{M}\Omega \text{ cm}$) supplied by an ultrapure water system (Aquapuri 5 series, Youngin Chromass).

Synthesis of transition-metal-alloyed Ga_2O_3 -capped EGaIn microparticles (LM@TM)

LM@TM microparticles were synthesized according to a modified literature procedure based on ultrasonication-assisted galvanic replacement.³⁰ Nickel(II) chloride hexahydrate ($\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$, 1,029.5 mg), iron(III) chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, 1,170.8 mg), cobalt(II) chloride hexahydrate ($\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, 1,030.6 mg), copper(II) chloride dihydrate ($\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, 738.4 mg), and zinc chloride (ZnCl_2 , 590.3 mg) were dissolved in ethanol, and the solution was adjusted to a

final volume of 43.3 mL. This afforded an equimolar five-metal precursor solution with a combined transition-metal concentration of 0.50 M (0.10 M for each metal). EGaIn (800 mg, containing 8.66 mmol of Ga) was then added to the precursor solution. The mixture was probe-sonicated in an ice bath at 35% amplitude for a cumulative sonication-on time of 10 min using a 3 mm-diameter (1/8 in.) tapered microtip with a pulse sequence of 7 s on and 3 s off.

Preparation of LM@TM-coated nickel fiber paper by dip coating

Following sonication, the LM@TM suspension was left undisturbed for 3 min to allow larger liquid-metal particles to settle. NFP substrates were then immersed in the upper portion of the suspension for 3 min without disturbing the sediment, withdrawn, and dried under vacuum at room temperature overnight.

Synthesis of SCCO by IPL photoforging

The LM@TM-coated NFP was subjected to IPL photoforging using a Photocura IPL system (PLT Co., Ltd.) equipped with a xenon flash lamp (First Light Technologies) emitting broadband UV–vis–NIR radiation. The lamp-to-sample distance was fixed at 1.5 cm, and a single shot was delivered at 400 V with a programmed pulse on-time of 20 ms. Unless otherwise stated, these irradiation conditions were used for all SCCO@NFP samples.

Structural characterizations

The morphology and microstructure of the synthesized SCCO electrodes were characterized by scanning electron microscopy (SEM, Regulus8230, Hitachi) and transmission electron microscopy (TEM, Titan, FEI). Selected-area electron diffraction (SAED) patterns were obtained during TEM

analysis to examine the crystallographic features of the SCCO samples. The fast Fourier transform (FFT) of the HRTEM images were performed using Gatan and ImageJ software. Elemental distributions were analyzed using energy-dispersive X-ray spectroscopy (EDS) combined with SEM or TEM measurements. The surface chemical states and elemental compositions were investigated by X-ray photoelectron spectroscopy (XPS, PHI 5000 VersaProbe, Ulvac-PHI) with an Al K α source ($h\nu = 1486.6$ eV). Raman spectroscopy was performed to examine oxide-related vibrational features and structural characteristics.

Electrochemical measurements

Electrochemical measurements (Bio-logic VSP Potentiostat) were performed in a standard three-electrode configuration using SCCO@NFP directly as the working electrode. A graphite rod and a Hg/HgO electrode were used as the counter and reference electrodes, respectively. All measurements were conducted in 1.0 M KOH electrolyte at room temperature. The measured potentials were converted to the reversible hydrogen electrode (RHE) scale according to the following equation:

$$E_{\text{RHE}} = E_{\text{Hg/HgO}} + E^{\circ}_{\text{Hg/HgO}} + 0.059 \times \text{pH}$$

Linear sweep voltammetry (LSV) was conducted at a scan rate of 2 mV s⁻¹ to evaluate HER and OER activities. HER polarization curves were recorded in the cathodic potential region, while OER polarization curves were measured in the anodic potential region. Commercial Pt/C and IrO₂ electrodes were used as benchmark catalysts for HER and OER, respectively. Bare NFP was also measured as a substrate control. All polarization curves were corrected with 80% iR compensation. Tafel slopes were calculated from the linear region of the Tafel plots using the equation:

$$\eta = b \log |j| + a$$

where η is the overpotential, b is the Tafel slope, and j is the current density. Long-term durability was evaluated by chronopotentiometry at fixed current densities. HER stability was measured at -100 mA cm^{-2} , and OER stability was measured at 100 mA cm^{-2} . The degradation rate was estimated from the voltage change during chronopotentiometric operation.

The electrochemically active surface area (ECSA) was estimated from the electrochemical double-layer capacitance (C_{dl}). Cyclic voltammetry was conducted in a non-Faradaic potential region at different scan rates. C_{dl} was obtained from the slope of the capacitive current density plotted as a function of scan rate, and the relative ECSA was compared based on the C_{dl} values.

Data availability

All data are available in the manuscript and its supplementary files. Source data are provided with this paper.

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Author Contributions

‡T.K. and Y.-J.R. contributed equally. T.K., J.-S.J., and T.O. conceptualized the project. T.K. and S.S. performed the microparticle processing. Y.-J.R. designed the IPL setup and optimized the processing parameters. All authors contributed to writing and approved the final manuscript.

Competing interests

The authors declare no competing interests.

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