Supplementary information for

Self-assembled Two-dimensional Copper Oxide Nanosheet Bundles as an efficient Oxygen Evolution Reaction (OER) electrocatalyst for water splitting application

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Materials: $CuSO_4.xH_2O$ (98%), NH_4OH (28-30% NH_3), KOH (>85%), H_3BO_3 (99.5%) and $Na_2B_4O_7.10H_2O$ (99.5%) were purchased from Sigma-Aldrich. The stainless steel substrates (Grade-304) were purchased from DAIHAN Scientific Co., Ltd, Korea. All the solutions were prepared using reagent-grade water (18 MΩ-cm resistivity).

Experimental: CuO nanosheet electrode films were synthesized by chemical bath deposition method on stainless substrate using a mixed solution of 0.1 M $CuSO_4$ and 1 M NH_4OH. The pH of the solution was kept constant at ~ 13 and pre-cleaned stainless steel substrate was immersed in the bath containing the solution, and the temperature was maintained at 80 ˚C for 6 hours. After the film deposition, the substrate was washed using DI water and then air-dried. The as-deposited film was annealed at 200 °C for 2 hours. A preliminary experiment was performed, and it was found that the as-deposited 2D CuO nanosheet film was optimized as an

electrocatalyst at 200 °C. The structural and chemical properties of the as-deposited and optimized CuO nanosheet films were studied using X-ray diffraction (XRD, Rigaku Smart Lab X-ray Diffractometer) with CuKα radiation (λ = 1.5418 Å) and X-ray photoelectron spectroscopy (XPS). Transmission electron microscope (TEM, JEOL-3000F) images and selected-area electron diffraction (SAED) patterns were captured at 300 kV. The morphology and chemical composition of the films were investigated using a field emission scanning electron microscope (FE-SEM, Hitachi SU8010) equipped with a Horiba Scientific energy dispersive spectrometer (EDS) analyzer. The electrochemical measurements were performed in a typical three-electrode system using an electrochemical workstation (Princeton Applied Research, VersaSTAT 3) at room temperature. The electrocatalytic OER measurements were performed in an O_2-saturated 1 M KOH electrolyte, using the 2D CuO nanosheet electrode with a geometric area of 1 × 1 cm^2 as the working electrode, a Pt wire as the counter electrode and a saturated calomel electrode (SCE) as the reference electrode. The potentials (E) in this work are referenced to a reversible hydrogen electrode (RHE) according to the Nernst equation in 1 M KOH ($E_{RHE} = E_{SCE} + 0.059\ pH + E^0_{SCE}$), where E^0_{SCE} is the standard potential of the SCE at 25 °C. Linear sweep voltammetry (LSV) was carried out by sweeping the potential from 1 V to 1.8 V vs. RHE at a scan rate of 10 mV/s, and several polarization curves were performed until the reproducibility of each measurement was obtained. To study the stability of the 2D CuO electrocatalyst, the chronoamperometry (CA) measurement was carried out at a constant potential of 1.58 V vs. RHE. The electrochemical impedance spectroscopy (EIS) was performed in the frequency range between 0.1 Hz and 1 MHz at 0.35 V with an AC potential amplitude of 5 mV.

Preparation of Borate buffer solution: The 0.2M borate buffer solution (pH~9) was prepared by mixing a 0.05M Na2B4O7.10H2O solution and a 0.2M H3BO3 solution in a volumetric ratio of 8:2.

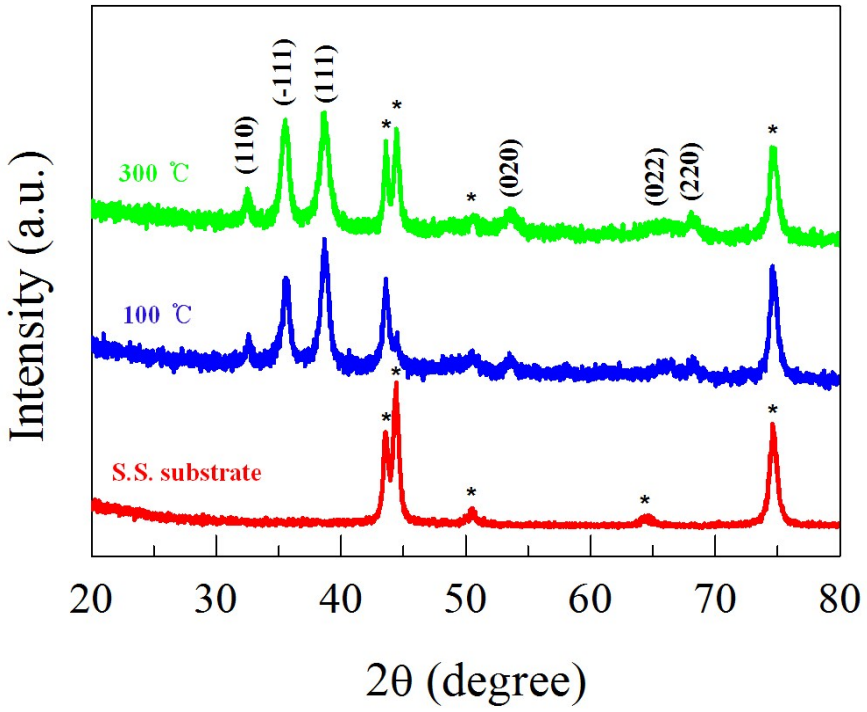

Fig. S1: X-ray diffraction patterns of 2D CuO nanosheet films annealed at 100 ^{0}C and 300 ^{0}C.

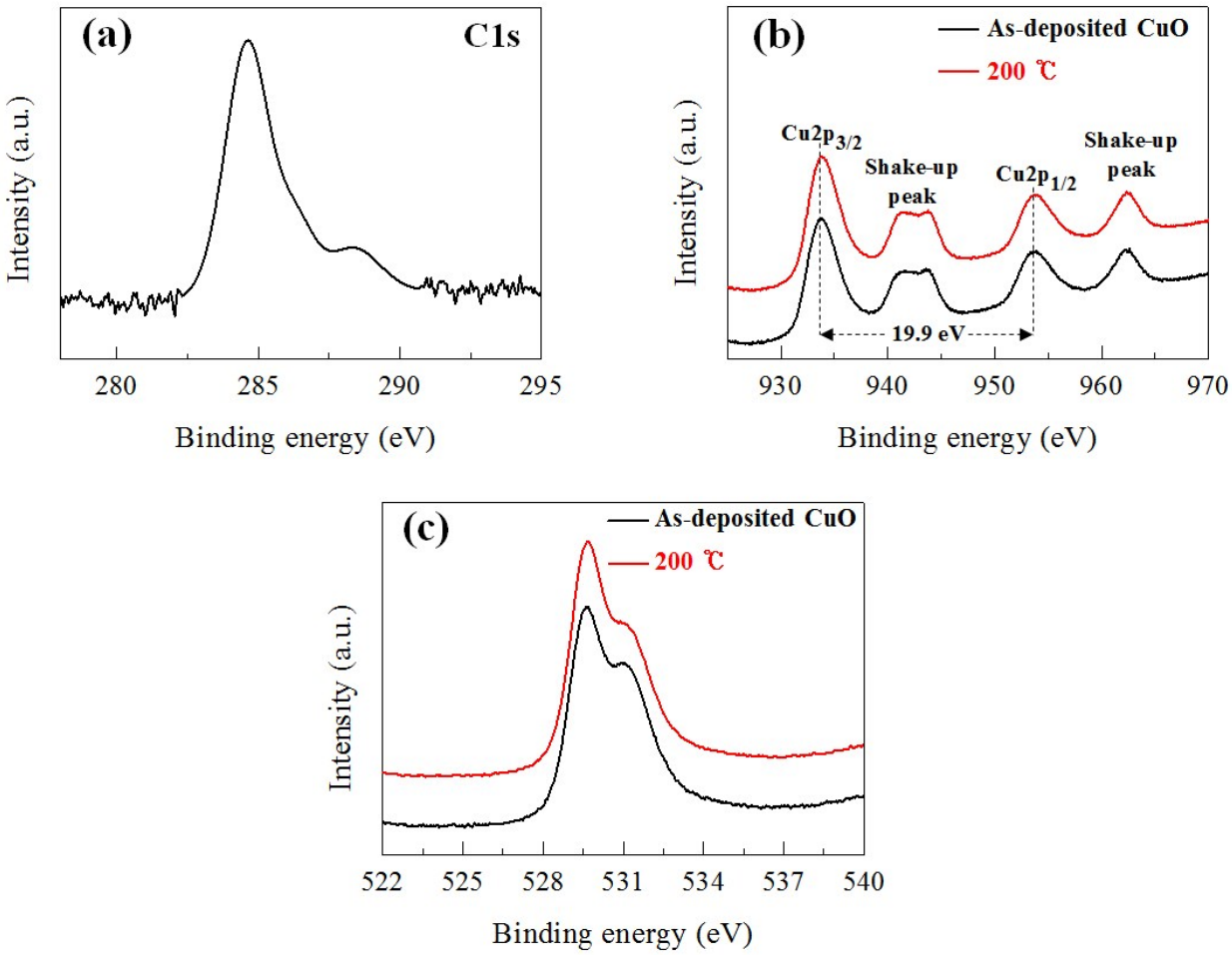

Fig. S2: (a) Core-level XPS of C1s, (b) core-level XPS spectra of Cu2p and (c) core-level XPS spectra of O1s for as-deposited and optimized annealed 2D CuO nanosheets films.

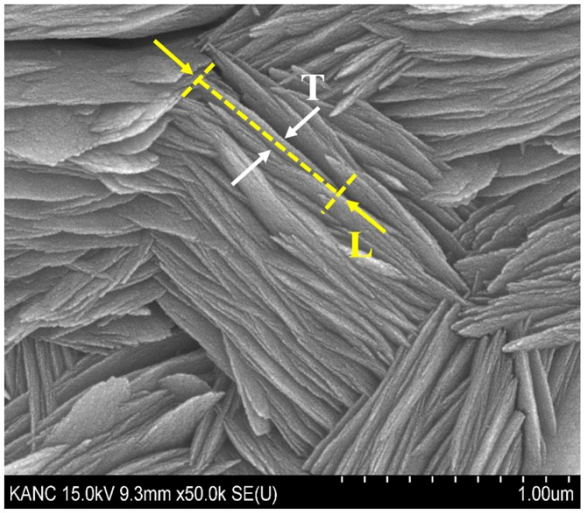

Fig. S3: Magnified SEM image of a 2D CuO nanosheet film.

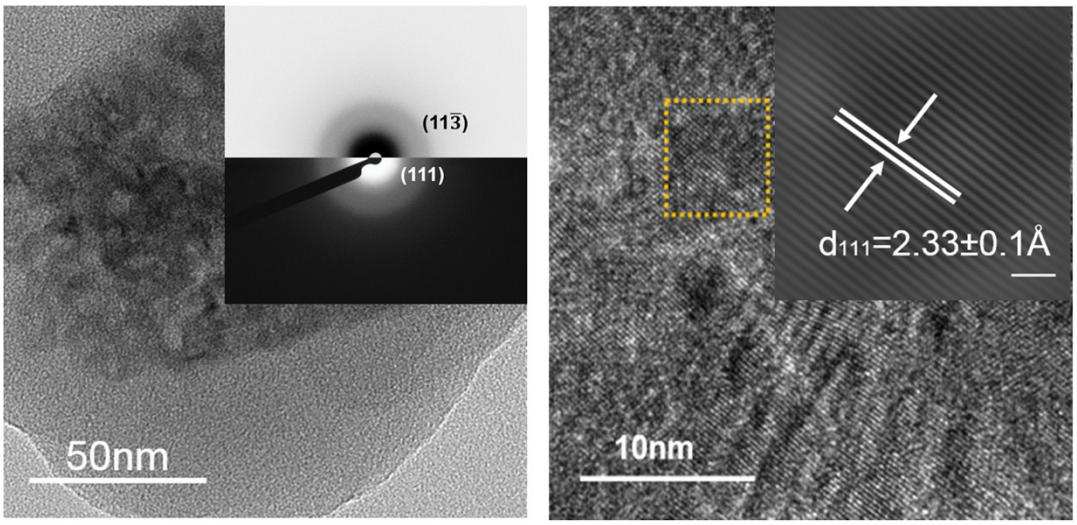

Fig. S4: TEM and HR-TEM images of a 2D CuO nanosheet film annealed at 300 ^{0}C. The insets show the corresponding SAED pattern and magnified HR-TEM image from the selected area of the HR-TEM image.

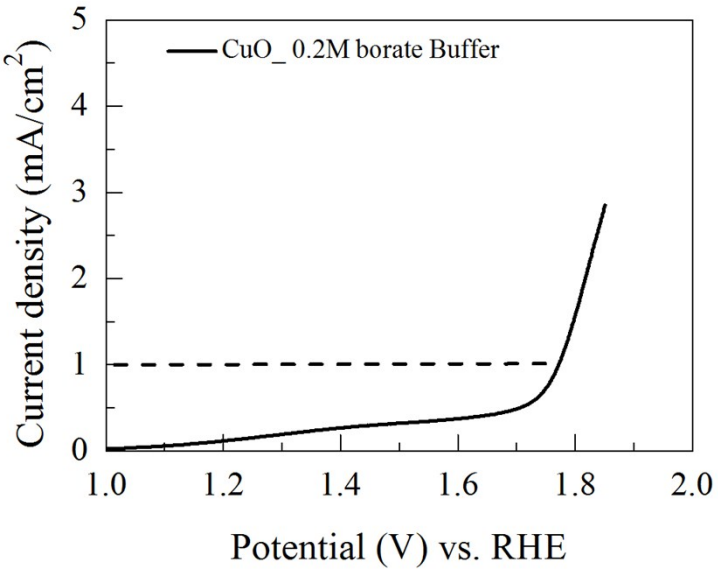

Fig. S5: Electrochemical OER linear sweep voltammetry (LSV) curve for the 2D CuO nanosheet electrode in a 0.2 M borate buffer solution.

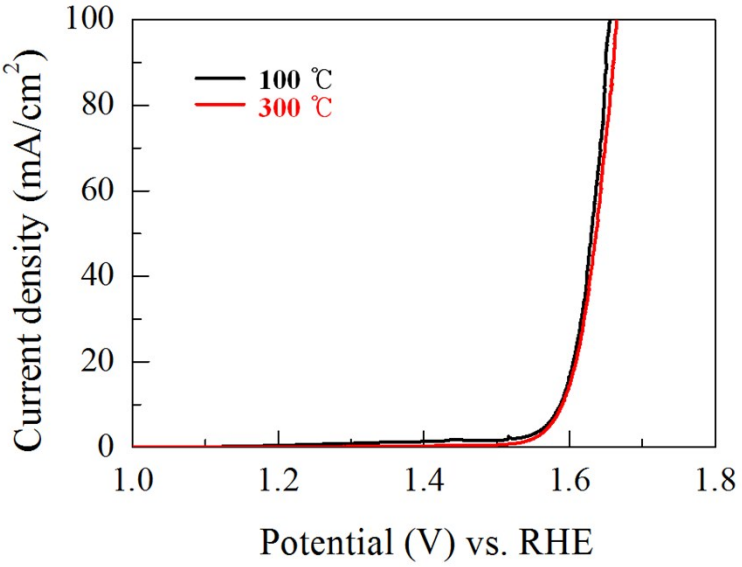

Fig. S6: Electrochemical OER linear sweep voltammetry (LSV) curves for the 2D CuO nanosheet electrodes annealed at 100 ^{0}C and 300 ^{0}C in a 1M KOH solution.

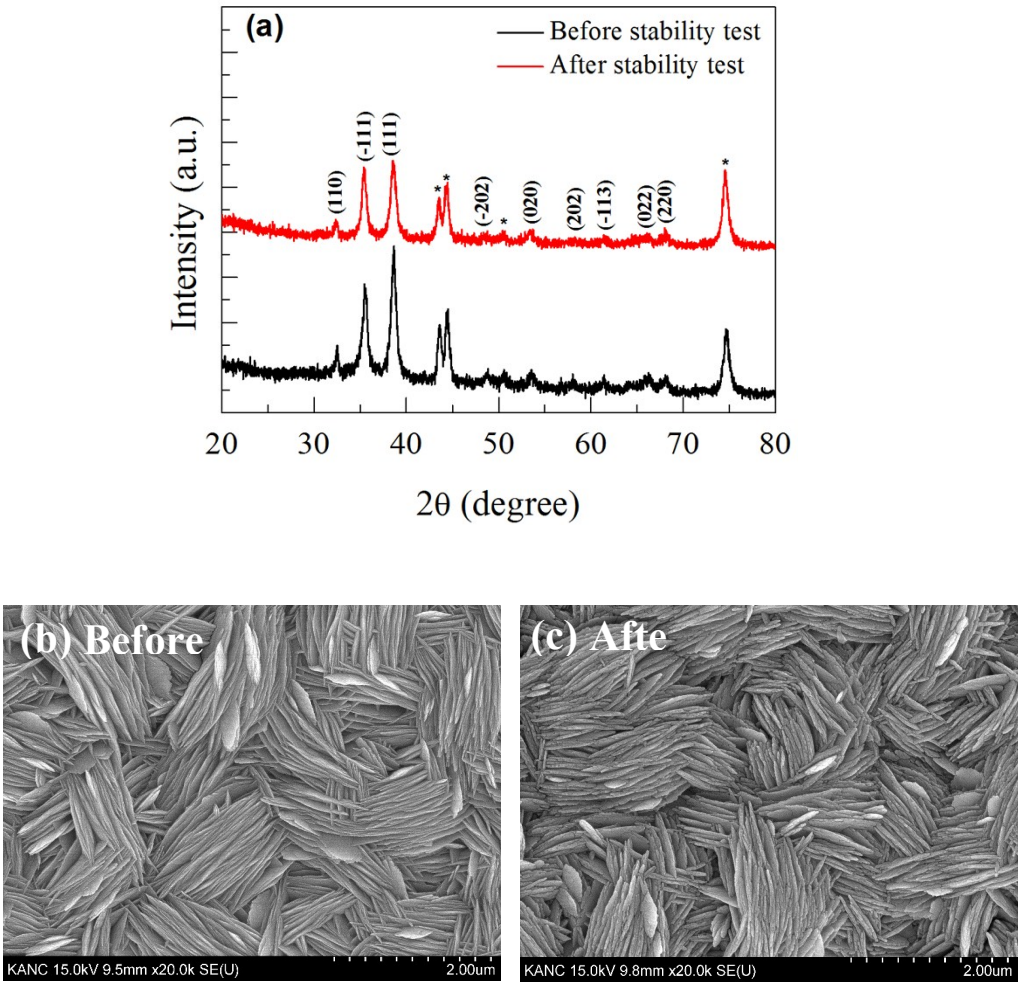

Fig. S7: (a) X-ray diffraction patterns and (b,c) SEM images of the 2D CuO nanosheet electrode before and after the stability test in a 1M KOH solution.

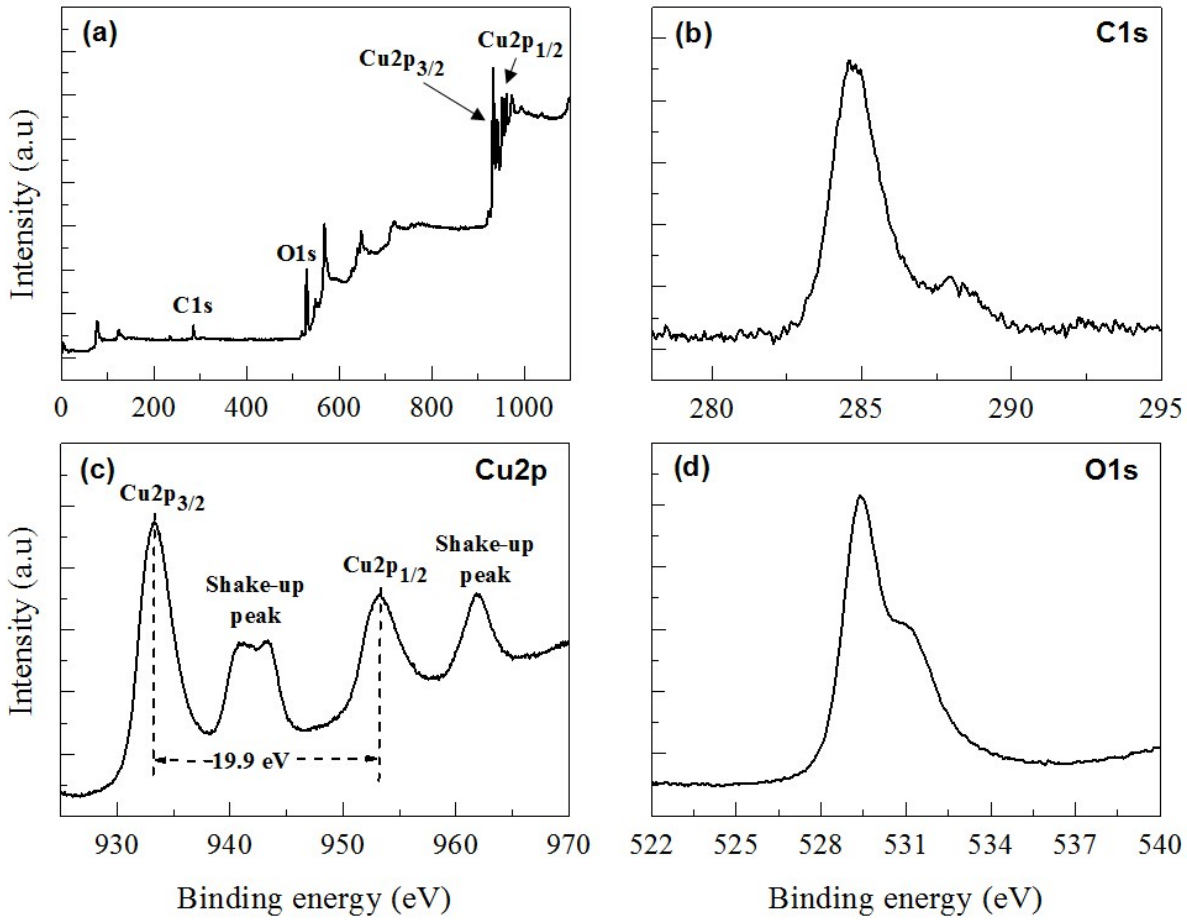

Fig. S8: (a) Full-scale XPS spectrum, (b) core-level XPS spectrum of C1s, (c) core-level XPS spectrum of Cu2p and (d) core-level XPS spectrum of O1s of a 2D CuO nanosheet electrode after the stability test in a 1M KOH solution.

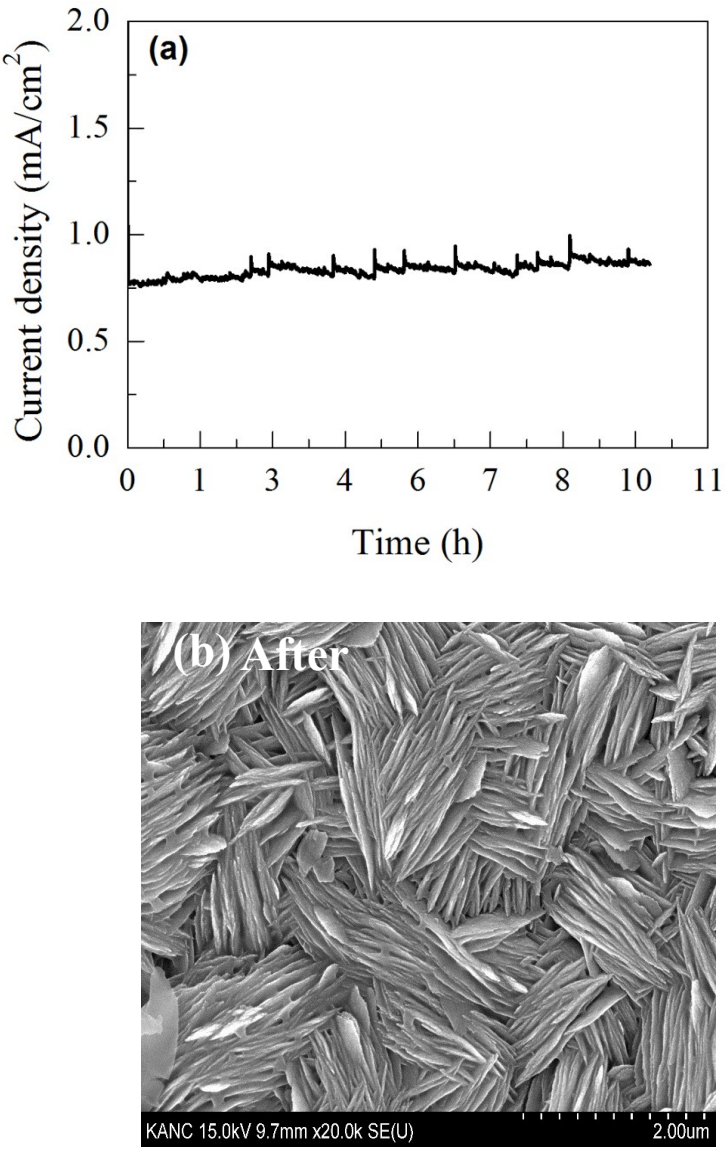

Fig. S9: (a) Chronoampetrometry stability test measured at 1.77 V/RHE and (b) SEM image after the stability measurement of the CuO nanosheet electrode in a 0.2M borate buffer solution.

Table S1: Compositional analysis of as-deposited and annealed 2D CuO nanosheets annealed at different annealing temperatures in the air atmosphere.

Sr. No.	Sample name	Cu (Atomic %)	O (Atomic %)
1	As-deposited CuO	47.09	52.91
2	100 ℃	51.38	48.62
3	200 ℃	51.19	48.81
4	300 ℃	51.32	48.68

Table S2: Electrocatalytic OER performance of Cu based oxide electrocatalysts in alkaline media.

Catalyst	Method	Electrolyte	Onset overpotential (mV)	Tafel slope (mV/dec)	Overpotential (η) @ 10 mA/cm²	Ref.
$Cu(OH)_2$/CuO	Anodization/ chemical oxidation	1 M KOH	350	76	417	[12]
Annealed CuO	Electro-deposition	1M KOH	360	61.4	580	[10]
CuO nanowire	hydrothermal	1 M KOH	340	54.5	-	[13]
CuO/Cu nanowire	electrodeposition	1M Na_2CO_3	380	90	580	[30]
CuO	Electro-deposition	1 M KOH	370	90	475	[14]
CuO	hydrothermal	0.1 M KOH	340	-	-	[15]
$Cu(OH)_2$	Electro-deposition	1M KOH	350	95	420	[31]
$Cu(OH)_2$	Precipitation	1M KOH	470	78	-	[11]
CuO nanoleaf	anodization	0.2 M Na_2CO_3	320	44	450	[16]
2D CuO nanosheet	Chemical bath deposition	1M KOH	250	59	350	This Work

[30] J. Du, Z. Chen, S. Ye, B. J. Wiley, and T. J. Meyer, *Angew. Chem. Int. Ed.,* 2015, **54**, 2073.

[31] X. Ma, X. Li, A. D. Jagadale, X. Hao, A. Abudula and G. Guan, *Int. Journal of Hydrogen Energy,* 2016, **41,** 14553.