Supporting information

Dynamically Controlled Growth of Cu-Mo-O Composite Nanosheets for Efficient Electrocatalytic Hydrogen Evolution

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Fig. S1 (a) XRD patterns of Cu powder reacted with $(NH_4)_6Mo_7O_{24}$ solution directly under stirring for 4 h, 8 h, 12 h, and 24 h; SEM images of Cu powder (b) and Cu powder reacted with $(NH_4)_6Mo_7O_{24}$ solution directly under stirring for 4 h (c), 8 h (d), 12 h (e), and 24 h (f).

Firstly, the perfect crystallization of metallic Cu with characteristic peaks (20) at 43.3°, 50.4° and 74.1° (PDF#. 04-0836). After the reaction for 4 h, the intensity of diffraction peaks of Cu decreased, but remained the main diffraction peaks and some new peaks at 29.6°, 36.4°, 42.3°, and 61.3° were ascribed to the (110), (111), (200) and (220) planes of cuprite Cu₂O (PDF#. 05-0667). With the reaction time increased to 8 h, the intensity of Cu peaks was weaken further and new peaks of 12.6°, 21.3°, 25.3°, 33.4°, 35.6°, 39.2°, 43.1° and 48.2° were consistent with (020), (-101), (031), (-141), (-112), (211), (-202) and (170) crystal face (PDF#. 36-0405), implying the formation of lindgrenite MO phase. After reaction for 24 h, the diffraction peak of Cu₂O and Cu were disappeared and new peaks at 17.8°, consistent with (011) crystal face of MO (PDF#. 36-0405). Therefore, the successful synthesis of CMO/CF in the presence of Cu₂O was due to excessive Cu on CF substrate. In addition, with the reaction time increasing, the morphology of the product changed as shown in **Fig. S1b-f**, and nanosheets which were appeared at the reaction for 8 h became more and more.



Fig. S2 MO precipitate obtained through Cu NWs/CF reacted with $(NH_4)_6Mo_7O_{24}$ solution under stirring condition.



Fig. S3 SEM images of MO powders (precipitation from the reaction of Cu NWs/CF and $(NH_4)_6Mo_7O_{24}$ solution under stirring condition).



Fig. S4 SEM images of CMOS/CF (CF reacted with (NH₄)₆Mo₇O₂₄ solution directly under stirring condition).



Fig. S5 SEM images of CMOS/CF under different stirring rate of (a, d) 300, (b, e) 600 and (c, f) 900 r/min.



Fig. S6 The thickness distribution of CMOS nanosheet under 600 r/min in 50 points.



Fig. S7 Energy dispersive spectrometer of CMOW (a) and CMOS (b) scratched from CMOW/CF and CMOS/CF.



Fig. S8 XPS survey spectra (a) and high-resolution XPS spectra of the Cu 2p (b), O and 1s (c) of Cu(OH)₂ NWs/CF, Cu NWs/CF, CMOW/CF and CMOS/CF.



Fig. S9 XPS survey spectra (a) and high-resolution XPS spectra of the Cu 2p (b), Mo 3d (c) and O 2p (d) of MO precipitate powder from the reaction of Cu NWs/CF and (NH₄)₆Mo₇O₂₄ under stirring condition.



Fig. S10 Selected area electron diffraction patterns of CMOW (a) and CMOS (b).



Fig. S11 XRD patterns (a) and polarization curves (b) of MO/CF, Cu_2O/CF and CMO/CF.



Fig. S12 Linear fitting of the capacitive currents of Cu(OH)₂ NWs/CF, Cu NWs/CF, CMOW/CF and CMOS/CF vs scan rates for HER.



Fig. 13 Polarization curves (a), linear fitting of the capacitive currents vs scan rates (b-d), and the capacitive currents (e) of CMOS/CF under different stirring rate for HER.



Fig. S14 Polarization curves corrected by electrochemically active area (a), Tafel plots (b), EIS Nyquist plots at 430 mV (c) of Cu(OH)₂ NWs/CF, Cu NWs/CF, CMOW/CF, CMOS/CF and (d) current-time plots with an overpotential of 300 mV for OER in 1 M KOH.



Fig. S15 Linear fitting of the capacitive currents of Cu(OH)₂ NWs/CF, Cu NWs/CF, CMOW/CF and CMOS/CF vs scan rates for OER.



Fig. S16 EIS Nyquist plots of full water splitting at 1.65 V.