Supporting Information

Synergistic copper nanoparticles and adjacent single atoms on biomass-derived N-doped carbon toward overall water splitting

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Fig. S1. (a) Ball and stick illustration of Cu$_3$(PyCA)$_3$·H$_2$O complex in single crystal form. (b) PXRD pattern and (c) FT-IR spectrum of Cu$_3$(PyCA)$_3$·H$_2$O. (d) TG curve of Cu$_3$(PyCA)$_3$·H$_2$O measured in N$_2$ atmosphere.
Fig. S2. (a) XRD patterns, (b) Raman spectra, (c) N$_2$ adsorption/desorption isotherms, and (d) the corresponding pore-size distribution curves of BDC-$x$ prepared at five different temperatures (i.e., 500, 600, 700, 800, and 900 °C).
Fig. S3. (a,b) SEM, and (c,d) TEM images of BDC-700.
Fig. S4. (a) The dependency of the N contents doped in BDNCs on the mass ratio between BDC-700 and melamine. (b) OER polarization curves of Ni foam, BDC-700, and BDNC with different N contents supported on Ni foams.
Fig. S5. Cu K-edge XANES spectra of Cu$_{1+n}$/BDNC, Cu foil, and CuPc samples.
Fig. S6. CV curves of (a) Ni foam, (b) Cu\textsubscript{1+n}/BDNC, (c) Cu\textsubscript{n}/BDNC, and (d) Cu\textsubscript{1}/BDNC at different scanning rates.
Fig. S7. PXRD patterns of Cu$_{1+n}$/BDNC with Cu contents of 0.5 and 2.3 wt.\%.
Fig. S8. (HR)TEM images of (a-c) Cu_{1+n}/BDNC (0.5 wt.% Cu) and (d-f) Cu_{1+n}/BDNC (2.3 wt.% Cu).
Fig. S9. (a) XPS survey, (b) Cu 2p, and (c) N 1s spectra of Cu1+n/BDNC (0.5 wt.% Cu).
Fig. S10. (a) XPS survey, (b) Cu 2p, and (c) N 1s spectra of Cu_{1+n}/BDNC (2.3 wt.% Cu).
Fig. S11. LSV curves of Cu$_{1+n}$/BDNC on GCE for electrochemical OER testing.
**Fig. S12.** (a) TEM, (b) HRTEM, and (c) HAADF-STEM images, (d) XPS survey, (e) Cu 2p, and (f) N 1s spectra of the used Cu$_{1+n}$/BDNC after 10 h OER test.