Reversible Switching of Single Atom to Diatom catalysts Supported on 1T'-MoS_2

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Abstract

Identifying a substrate that provides strong anchoring of two different types of single atom catalysts (SACs) while enabling the dynamic re-configuration of SACs into heteronuclear dual atom catalyst (DAC) benefits the electrochemical synthesis, but remains relatively unexplored. In this study, we demonstrate that the electrochemical desulfurization of MoS_2 produces abundant 1 T' domains that facilitate the high loading of Cu (7.9 wt%) and Pt (6.7 wt%) to form DACs. By using operando X-ray absorption spectroscopy and first-principles calculations, we revealed the dynamic, reversible configuration between single atom state (Pt and Cu) and heteronuclear DAC state (Cu-Pt) by applying a voltage near hydrogen evolution reaction during electrochemical reactions. As a proof of principle, we demonstrate that the Cu-Pt DACs induced by electric field showed superior performance for the selective hydrogenation of alkynes compared to mono-elemental Cu or Pt SACs. Our research highlights that vacancy-enriched 1T'-MoS_2 serves as a useful template for the high loading of SACs—essential for accessing the electric bias-induced reconfiguration of catalyst structure, thus affording a new route for the on-demand fabrication of intermetallic single atom catalysts in electrosynthesis.

References

[1] Jianhua Wu, Kian Ping Loh et. Al. * Electric Bias-induced Reversible Configuration of Single and Heteronuclear Dual-Atom Catalysts on 1T'-MoS_2, ***Nat Nanotechnology*** **(**2025).

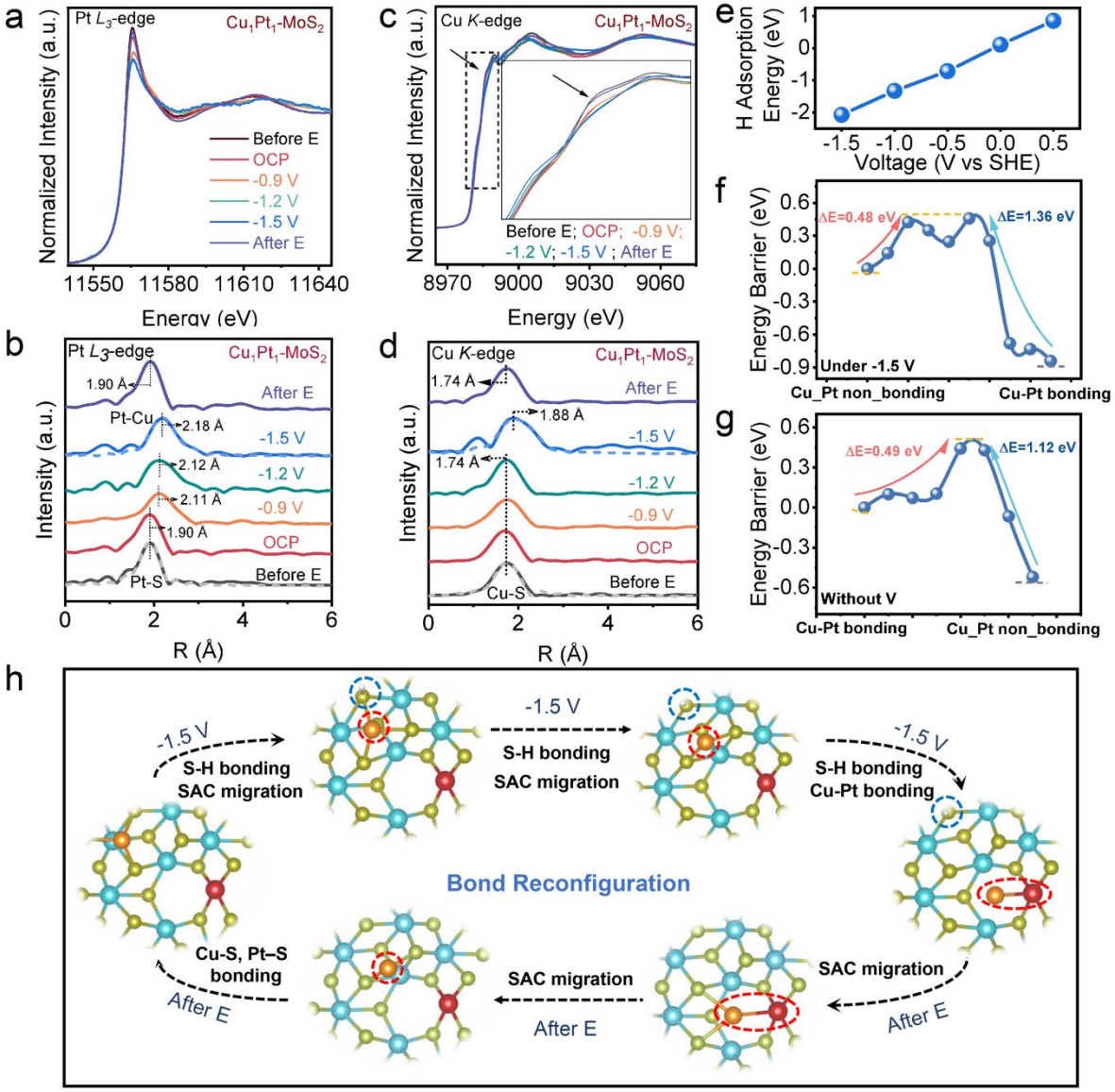

Fig 1. Operando-XAS of Cu_1Pt_1-MoS_2 before and after application of electrical bias. a) Operando Pt L_3-edge XANES spectra of Cu_1Pt_1-MoS_2 recorded at different applied voltages. b) Corresponding FT-EXAFS spectra from (a). c) Operando Cu *K*-edge XANES spectra of Cu_1Pt_1-MoS_2 recorded at different applied voltages. d) Corresponding FT-EXAFS spectra from (c). e) H binding energy on S dangling bond to form SH under different voltage. f) Calculated energy barrier of single atoms diffusing to form Cu-Pt bonding under negative voltage, with formation of S-H bond. g) Calculated energy barrier of Cu-Pt un-bonding after bias off, with H desorbed from S. h) Scheme showing the time-evolution of the bias-driven bond reconfiguration; the bonding and un-bonding of Cu-Pt SACs can be reversibly switched by bias "on" or "off". The green, blue, orange, red and gray balls represent the S, Mo, Cu, Pt, H atom, respectively.