In situ Time-resolved X-ray Absorption Spectroscopy Unveils Partial Re-Oxidation of Tellurium Cluster for Prolonged Lifespan in Hydrogen Evolution

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Abstract

Efficient and long-lasting electrocatalysts are one of the key factors determining their large-scale commercial viability.[1] Although fundamentals in deactivation and regeneration of electrocatalyst are crucial to understand and sustain durable activity, little has been conducted to metalloids rather than metal-derived ones. Herein, by virtue of *in situ* seconds-resolved X-ray absorption spectroscopy we discovered the chemical evolution in deactivation-regeneration cycles of tellurium clusters supported by nitrogen-doped carbon (termed Te-ACs@NC) as high-performance electrocatalyst in hydrogen evolution reaction (HER) (Figure 1). Through *in situ* electrochemical reduction, Te-ACs@NC that was deactivated due to surface phase transitions in a previous HER process, was re-activated and regenerated for the next run, where partially oxidized Te was found surprisingly to perform better than its nonoxidized state. After 10 consecutive deactivation-regeneration cycles for 480 hs, the Te-ACs@NC retained 85% of its initial catalytic activity. Theoretical studies suggest that local oxidation modulates the electronic distribution within individual Te clusters, to optimize adsorption energy of water molecules and reduce dissociation energy. This study provides fundamental insights into rarely explored metalloid cluster catalysts during deactivation and regeneration, and will assist future design and development of supported catalysts of high activity and long durability.

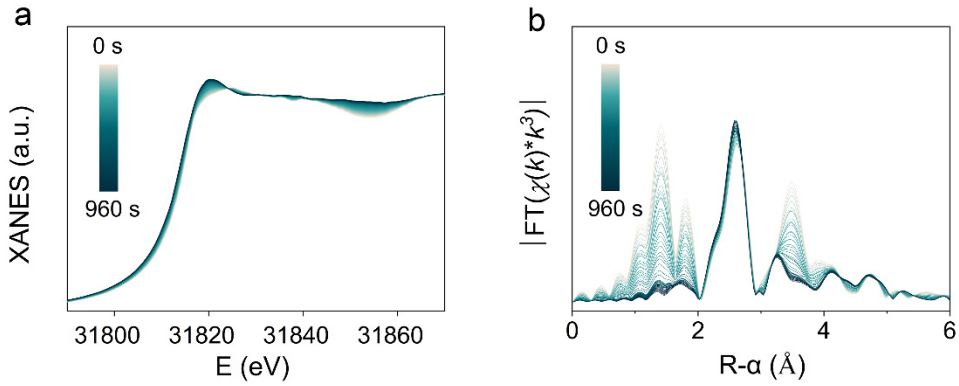

Figure 1. (a) *In situ* time-resolved Te K-edge XANES spectra and (b) the corresponding time-resolved EXAFS spectra under *in situ* electrochemical reduction at -1.1 V vs. RHE.

Reference

1 Liu, S. *et al.* Ultrastable Au nanoparticles on titania through an encapsulation strategy under oxidative atmosphere. *Nat. Commun.* **10**, 5790 (2019).