Cost-effective Synthesis of Cu_3N Photocathodes for Solar Energy Conversion Applications

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Abstract:

Converting sunlight into synthetic liquid solar fuels or electricity using photoelectrochemical (PEC) and photovoltaic (PV) routes, respectively, is a leading approach for addressing rising global energy demand. Cu_3N is a promising earth-abundant photocathode material due to its ideal 1.8 eV bandgap, high absorption coefficient, and good charge carrier mobility; however, no synthesis strategies have yet been reported that demonstrate photoactive material. Here, by virtue of in-situ X-ray diffraction measurements during nitridation of metallic Cu films in a NH_3:O_2 atmosphere, we developed a new method of sequential heating/cooling cycles that significantly improved the crystal and microstructural qualities of Cu_3N and yielded an appreciable photocurrent, for the first time.

Introduction

The realization of sunlight as a primary energy source, requires photons to be captured, converted, and stored in a cost-effective fashion, which requires the development of novel earth-abundant photoactive materials. Among these materials, Cu_3N is of particular interest due to its narrow 1.8 eV bandgap, high absorption coefficient of 10^5 cm^{-1} above 2 eV, high charge carrier mobilities (up to 200 cm^2/V.s), and bipolar self-doping.

Outlook

- By virtue of in-situ XRD measurements we found that Cu_3N was only formed at a temperature of 400°C during cooling from 550°C.
- We developed a new saw-tooth nitridation technique, that improved the overall film quality and yielded photoactive Cu_3N films.
- Cu_3N exhibited a reasonable photocurrent, which was progressively increased with time during the stability test.

Team

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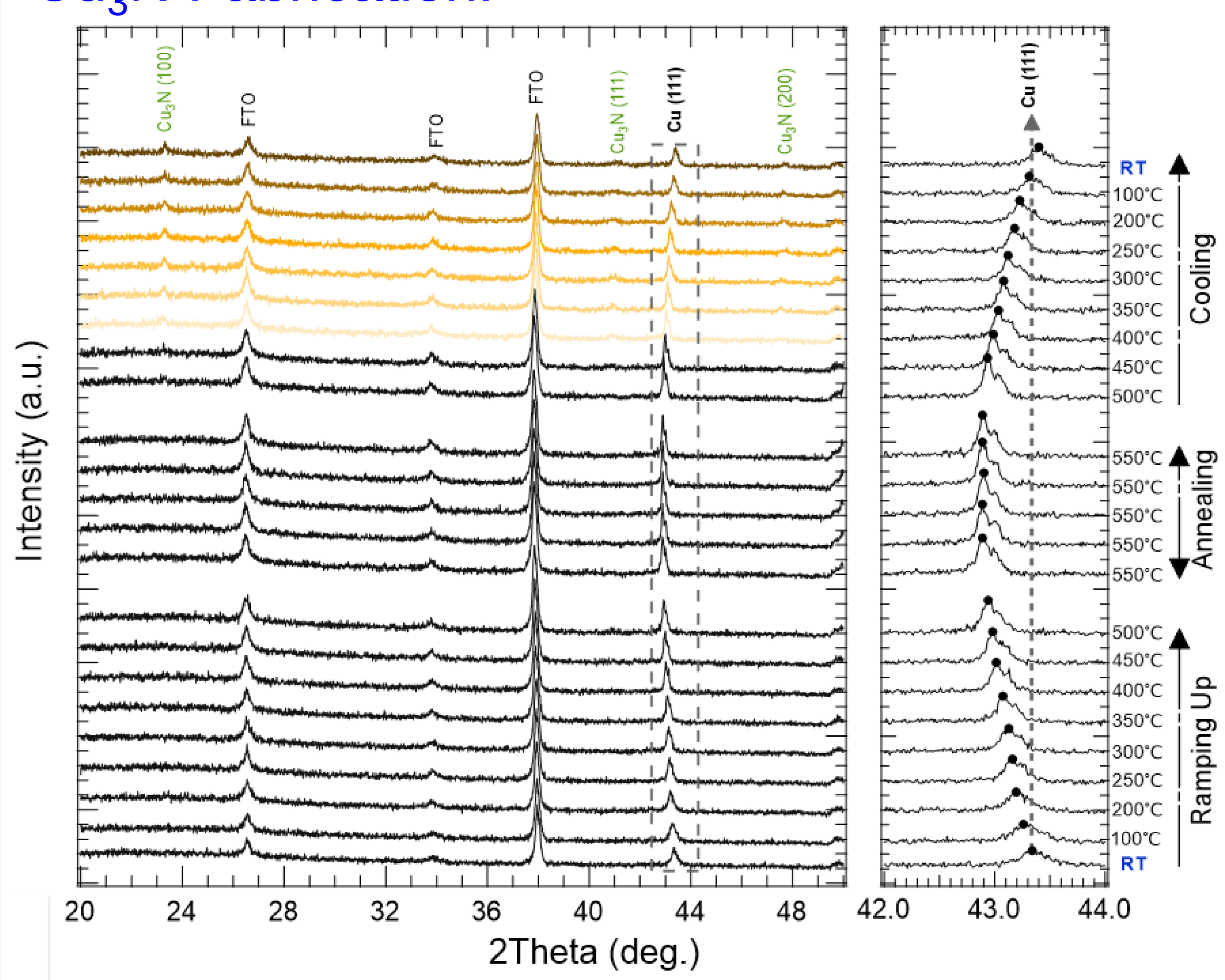
Acknowledgement

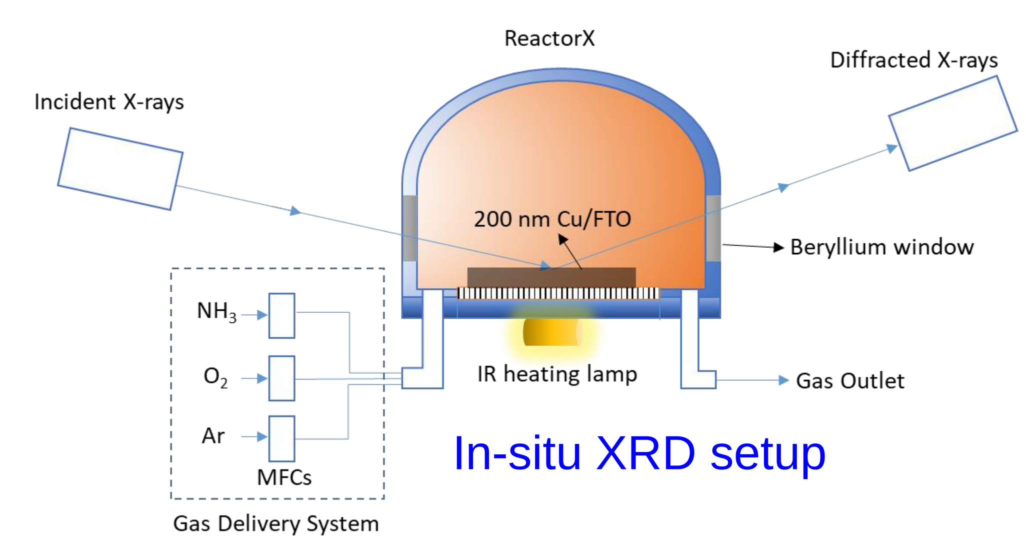
This work performed by the Joint Center for Artificial Photosynthesis, a DOE Energy Innovation Hub, supported through the Office of Science of U.S. Department of Energy under Award Number DE-SC0004993.

Results, Highlights, and Accomplishments

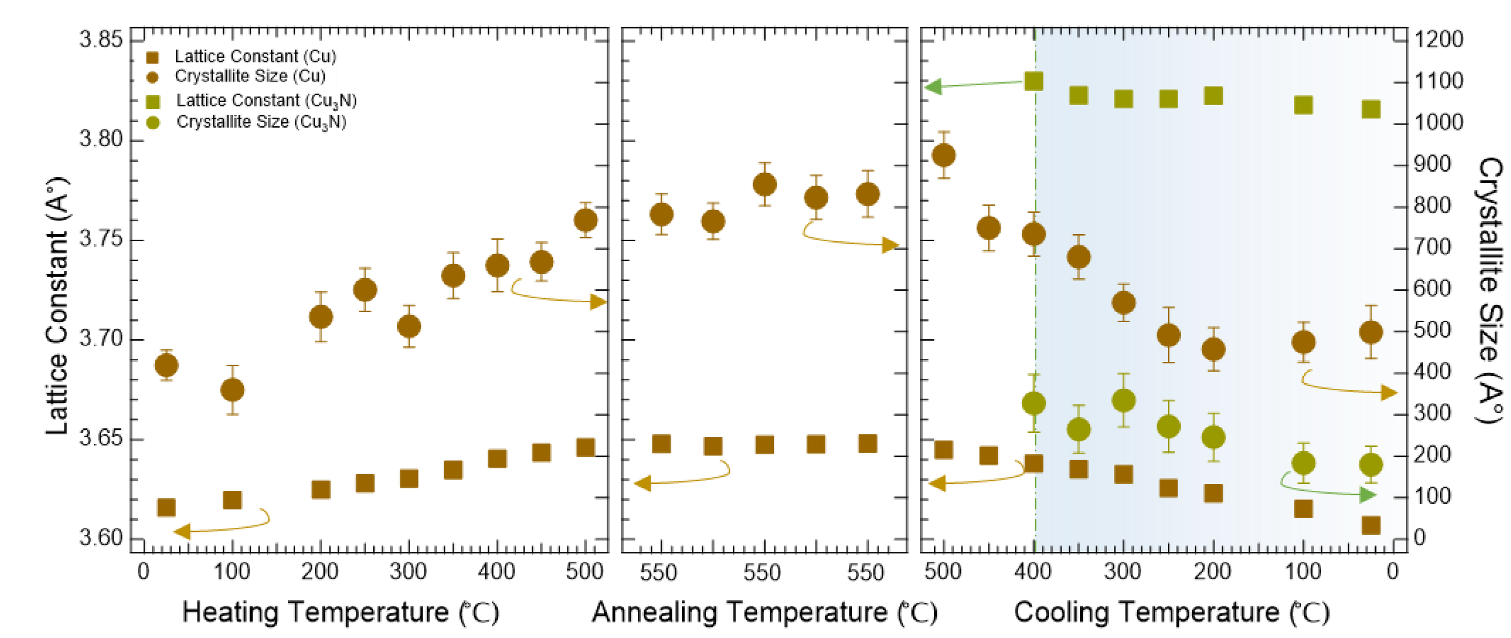
Cu_3N Fabrication:



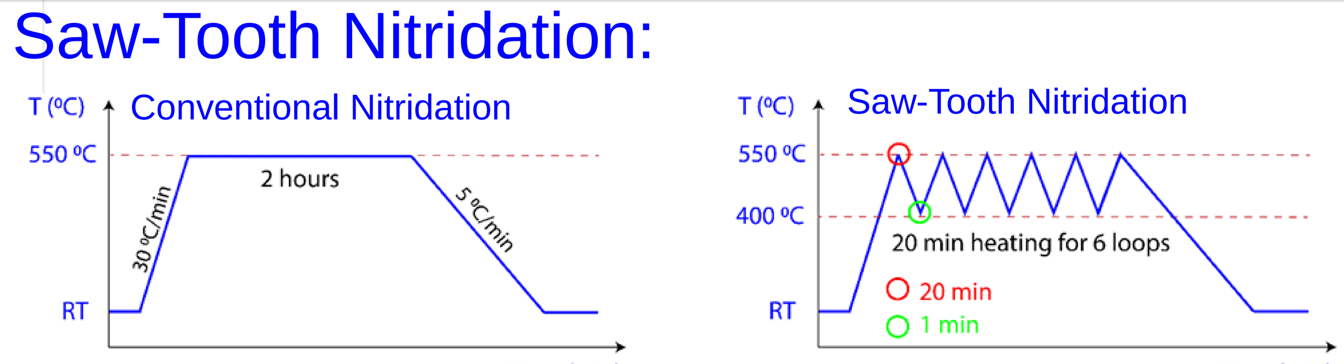
Temperature-dependent crystalline-phase evolutions

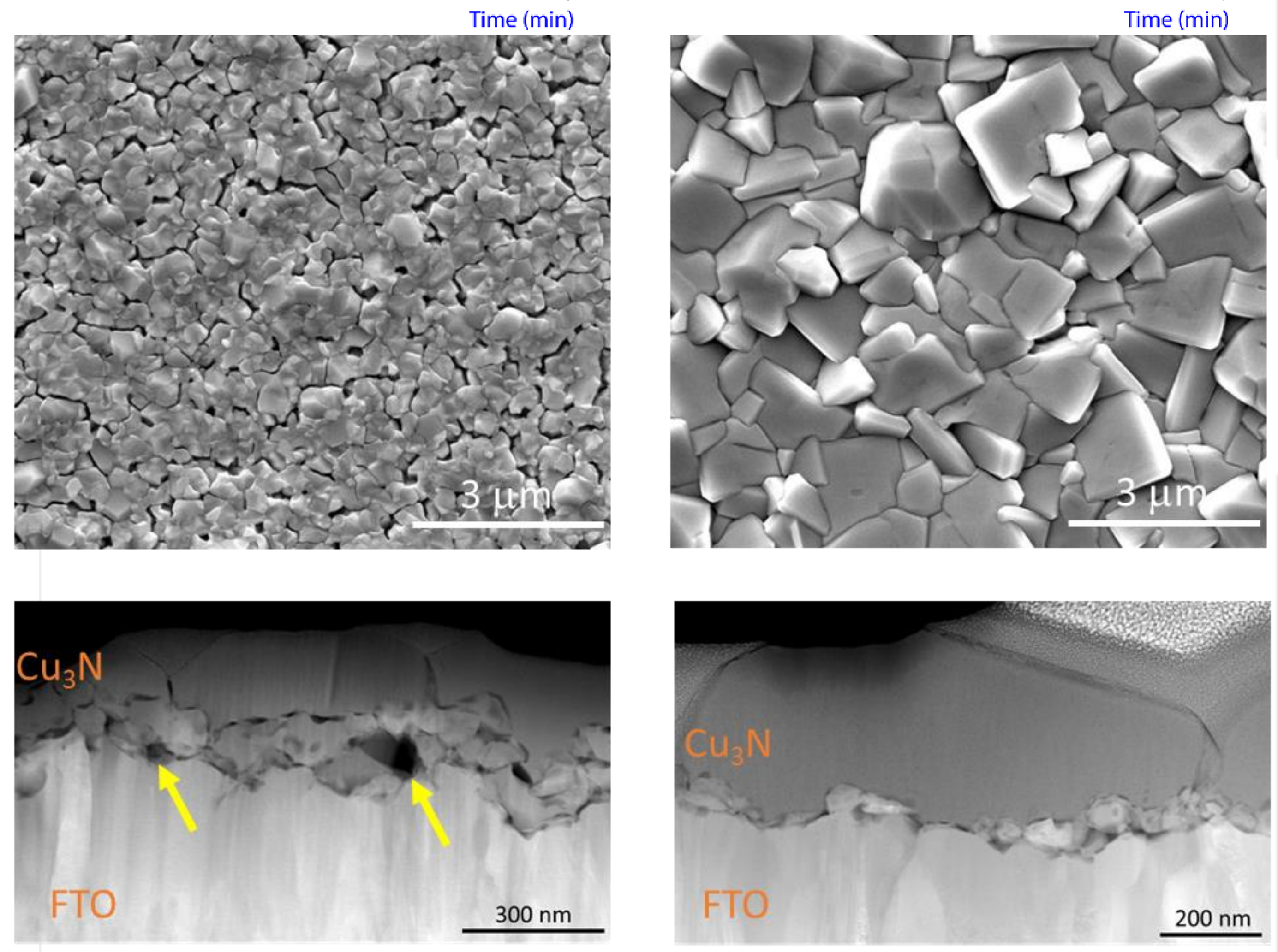


In-situ XRD setup

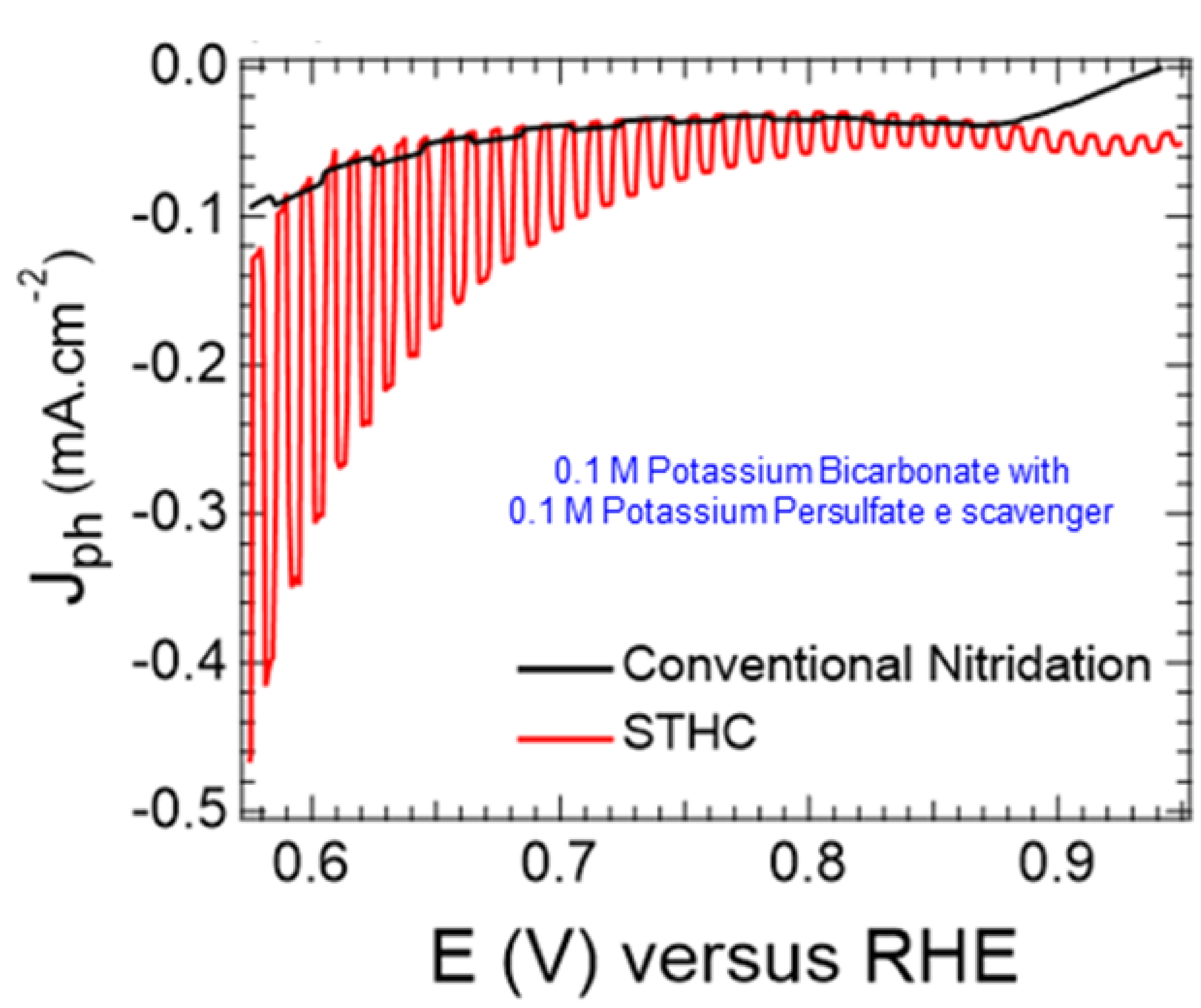


Saw-Tooth Nitridation:



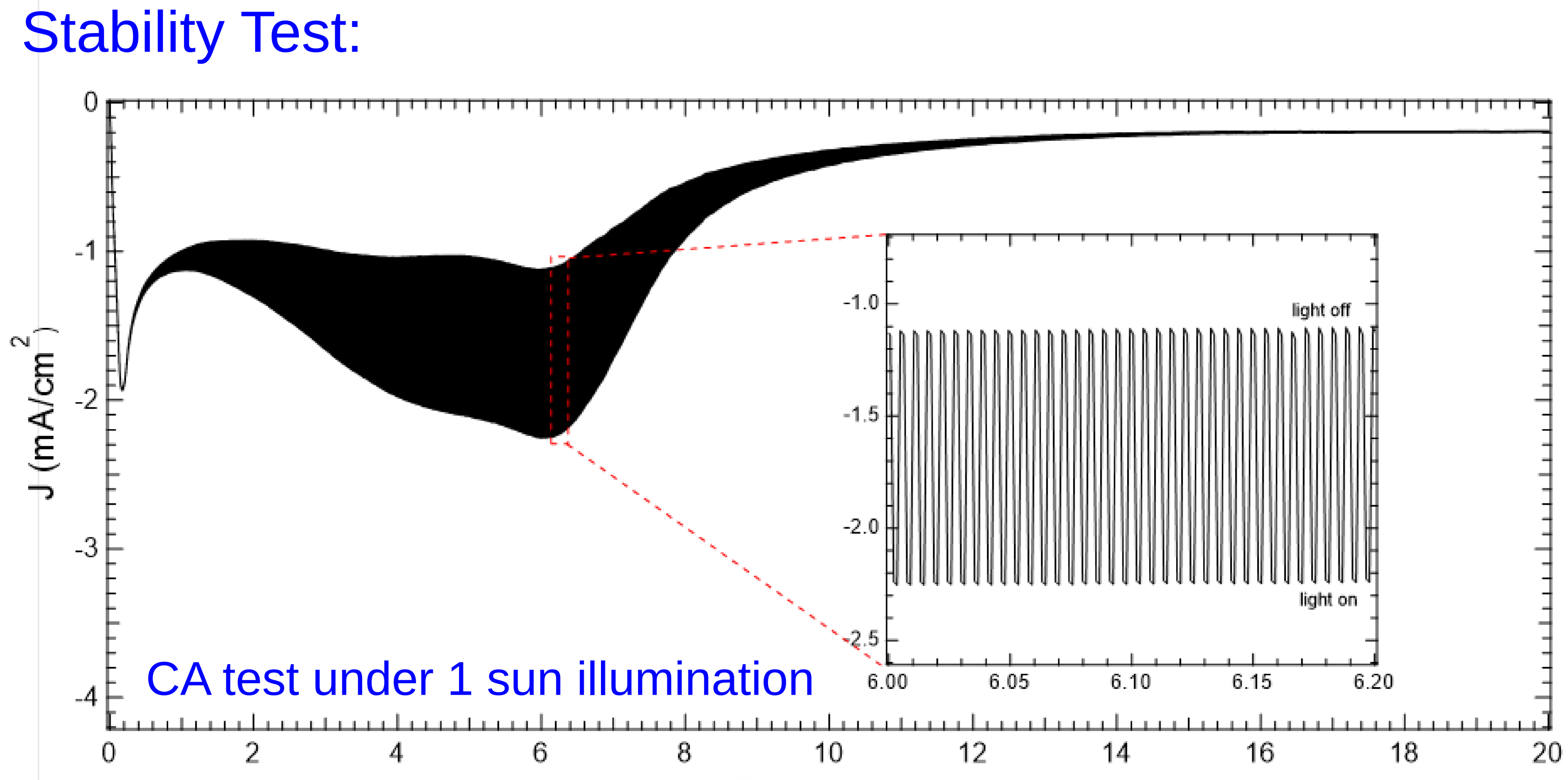


PEC Performance:



Cu_3N grown by conventional nitridation has no photocurrent
Cu_3N grown by STHC exhibited a photoresponse for the first time

Stability Test:



Surface morphology after 6 hrs in the electrolyte