Improving durability of III-V based PEC electrode: Atomistic insight from theory and experiments

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Photoelectrochemical (PEC) cells based on III-V semiconductor photocathodes have demonstrated highly efficient conversion of solar energy to hydrogen fuel.[1] However, photocorrosion of the electrode in electrolyte solution remains a significant challenge. Surface and solvent treatments offer the possibility for improving durability while retaining high solar conversion efficiency. Developing a coherent strategy for device improvement relies on a fundamental knowledge of the complex chemistry active at the electrode-electrolyte interface, which remains poorly understood.

Recently, it was reported that nitrogen ion bombardment on $GaInP_2$ improves its durability significantly with an acceptable level of sacrifice on the solar-to-hydrogen conversion efficiency (see Fig 1).[2] In order to understand the mechanism of electrode stabilization, we have started series of experimental and theoretical studies on III-V surface, on its interfaces with electrolyte, and on the chemical environment of nitrogen implanted in the III-V electrode.

At the presentation, we will first discuss on the microscopic properties of pristine/oxidized/hydroxidized III-V semiconductor surfaces,[3] and the chemical property of III-V/electrolytes interfaces studied by first-principles simulations.[4] We will then discuss on the chemical environment of nitrogen in the III-V electrode measured by various experimental methods, particularly X-ray based spectroscopy such as X-ray absorption/emission spectrum, and its interpretation based on first-principles computer simulations (see Fig 2).

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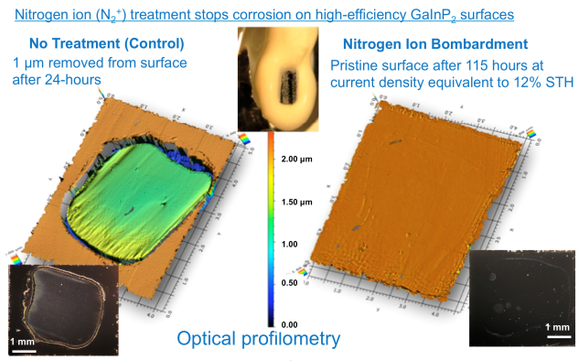

Fig 1: Enhanced durability of GaInP2 electrode using nitrogen bombardment demonstrated by Turner and Deutsch.[2]

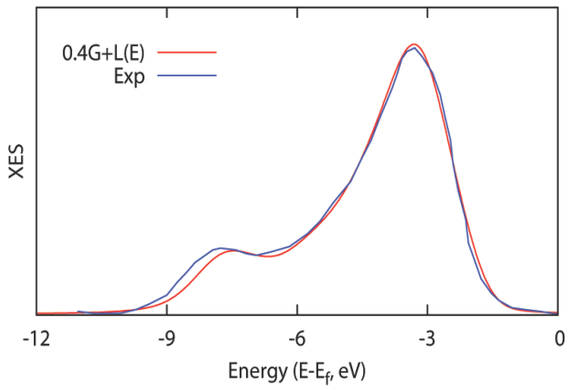

Fig 2: N K edge X-ray emission spectrum of GaN compound. Exp (blue line) is an experimental one from ref [5]. "0.4G+L(E)" is a theoretical spectrum based on Density Functional Theory with Gaussian broadening (σ=0.4eV) and energy dependent lifetime broadening (Lorentzian type) whose parameters were determined in this work. The X-ray code developed by Prendergast was used.[6] The same calculation procedure is used to investigate on the chemical environment of nitrogen impurity in $GaInP_2$ electrode.