Structure–Property Relationship of Oxygen-Doped Two-Dimensional Gallium Selenide for Efficient Water Splitting

Ephrem G. Demissie,[1] Wai Kit Tang,[2] Chi-Kit (Andy) Siu*,[1]

[1] *Department of Chemistry, City University of Hong Kong, Hong Kong SAR, P. R. China*
[2] *Department of Chemistry, Universiti Malaya, Malaysia*
chiksiu@cityu.edu.hk

Two-dimensional gallium selenide (GaSe) is a potential candidate for photocatalytic water splitting. However, its inert surface and wide bandgap limit its catalytic efficiency for hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). This talk summarizes our recent density functional theory (DFT) investigations regarding the effects of substitutional oxygen doping on electronic properties and photocatalytic activities of a single-layer GaSe.[1,2] In general, O doping reduces bandgap from 2.78 eV for pristine GaSe to 2.09 – 2.21 eV for $GaSe_{1-x}O_x$ (with x up to 22%) mainly due to lowering of conduction band minima. Applying tensile strain (up to 1%) further reduces the bandgap to 1.95 – 2.05 eV, further enabling visible-light absorption. The catalytic performance of $GaSe_{1-x}O_x$ strongly depends on the atomic configuration of O dopants. For HER,[1] more localized dopant arrangement significantly enhances hydrogen adsorption efficiency; this improvement is attributed to the weakening of Ga−O covalent interactions, which strengthens H binding. Manipulating the atomic arrangement of O dopants is also essential for enhancing the performance of OER.[2] When O dopants are separated by at least one -Ga-Se-Ga- unit, the reaction is limited by strong adsorption of the O* intermediate. By contrast, Ga atoms bonded to three O dopants act as optimal active sites, reducing the overpotential from 1.52 V for pristine GaSe to 0.38 V for the best performing $GaSe_{1-x}O_x$ (x = 22%), which is comparable to many state-of-the-art metal-based catalysts. This study highlights the importance of controlling the atomic configuration of O dopants to optimize the catalytic activity and bandgap of $GaSe_{1-x}O_x$, offering a pathway to engineer efficient photocatalysts for solar-driven water splitting.

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References

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