Abstract

In this thesis, we have studied the electronic and optical properties of functionalized $g-C_3N_4$ along with possible photocatalytic reaction mechanisms mainly for hydrogen evolution reaction (HER) and oxygen evolution reaction (OER) using Density Functional Theory. Starting from the study of monolayer $g-C_3N_4$ and its quantum confinement from bulk-phase, functionalization of $g-C_3N_4$ by doping of non-metals (B, O, P)/loading of metals (Co, Fe, Ag) along with suitable doping-loading (B-Co, O-Fe, P-Ag) have been studied. A system with suitable band edges was screened to perform overall water splitting. Next, functionalization of $g-C_3N_4$ by the formation of metal-semiconductor heterojunction using Cobalt-boride (CoB) in 0D and 2D form has been considered to understand the role of dimensionality in charge transfer across the interface. Further, the semiconductor-semiconductor heterostructure comprising of MXene (Hf_2CO_2) over $g-C_3N_4$ provides insights into band bending in OER/HER. Structural modification in the pristine $g-C_3N_4$ by different stacking of bilayer and Li intercalation in the most favorable bilayer have also been investigated. An elaborative study of charge transfer over conventional and simultaneous water-splitting reactions using climbing image nudge elastic band (CI-NEB) shows it as a potential photocatalyst. This thesis also unravels the other photocatalytic reaction mechanisms like carbon dioxide reduction reaction and nitrogen reduction reaction over the single atom catalyst ($Co-g-C_3N_4$) with possible reaction intermediates and reaction pathways to determine the most feasible reaction. Overall, this thesis describes the role of different functionalization over photocatalytic activities and defines strategies to engineer band gap of wide band gap semiconductor $g-C_3N_4$, to improve the photocatalytic performance from the variation at the atomistic level.