Preface

'The important thing is to never stop questioning"
Albert Einstein

The phenomenal discovery of the *Honda-Fujishima effect* in 1972 by Prof. Kenichi Honda and his disciple Prof. Akira Fujishima has inspired a half-century of researchers to study photocatalytic water splitting. Their work introduced, the potential of titanium dioxide electrodes for water decomposition on exposure to the visible spectrum of light, to the global scientific community. Thus, initiating a breakthrough in the field of photocatalysis, and photocatalytic materials. A useful method developed for water photolysis, for the first time, revolutionized the self-cleaning properties and unraveled the effectiveness of light in electrochemical systems.

The presence of high visible light absorbance, an effective charge carrier separation, and band edges straddling reduction-oxidation potential are essential prerequisites possessed by photocatalysts. These properties reflect on the generation of photo-induced charge carriers, their rapid migration, and surface activation for corresponding reactions. Investigation of photocatalytic properties on two-dimensional (2D) semiconductors and the role of functionalization over efficiency have been explored utilizing *Kohn-Sham* equation-based Density Functional Theory, in this thesis. Hydrogen evolution reaction, and oxygen evolution reaction study over the photocatalyst followed by carbon-dioxide reduction reaction, and nitrogen reduction reaction have been simulated to understand the feasibility of reaction pathways. Potential candidates demonstrating a low reaction barrier have been discovered for efficient energy conversion applications.

This thesis includes seven chapters, with four working chapters. Initially, an understanding of photocatalysis along with photocatalytic materials was established followed by computational methods utilized to probe their physical-chemical properties. Work done in this thesis begins with the investigation of graphitic carbon nitride ($g - C_3N_4$), and the effect of metal/non-metal loading/doping, loading, and doping on the photocatalytic efficiency. The role of 0D/2D-2D metal-semiconductor, and 2D/2D semiconductor-semiconductor heterojunction in improving the reaction feasibility has been uncovered in the following chapter. Further, the analysis of both pristine and modified $g-C_3N_4$-bilayer have been studied. An effective photocatalytic material by investigation of multiple pathways for CO_2, and N_2 reduction, demonstrated its applications in the realm of energy conversion. At last, the conclusion was marked by the effectiveness of photocatalytic materials in the present scenario, and future prospects were discussed that would result from the current work.