PREFACE

Graphitic carbon nitride (gCN), a metal-free, two-dimensional (2D) material, has garnered significant interest due to its high chemical/mechanical stability, tunable molecular structure, catalytic efficacy, environmental sustainability, and easy synthesis. From a structural perspective, gCN features a structure close to graphene with a honeycomb configuration with s-triazine or tri-s-triazine aromatic heterocycle as the fundamental unit. The sp^2 hybridization of carbon and nitrogen atoms in the aromatic ring of tri-s-triazine gCN establishes a delocalized π-conjugated system with C-N layers linked by weak van der Waals forces. Because of the π-conjugated system, abundant C and N sites, and tunable band gap, gCN swiftly emerged as a prominent green catalyst in the domain of photo-catalysis, including CO_2 capture and reduction, photocatalytic degradation, and photocatalytic water splitting reactions. Nonetheless, the application of gCN in the electrochemical field has not been explored much. Its restricted use is due to its low electrical conductivity, leading to sluggish charge transfer. Another problem is its bulk structure, which is mainly obtained from the thermal pyrolysis of N and C-rich precursors like melamine. The bulk structure offers a limited surface area and poor mass transfer, offering a suppressed electrochemical response. Its low dispersibility in most of the solvents and high N content are a few other problems that make bulk gCN an ineffective choice for electrochemical processes. However, the ease of surface functionalization and its ability to interact with various metals offer tremendous venues for tailoring its composition and structure to tune the physicochemical properties of the bulk gCN as per the electrochemist's need.

Along the same line, this thesis is focused on overcoming the aforementioned drawbacks associated with gCN and utilizing it as an electrocatalyst by tailoring its morphology, structure, and composition. The thesis revolves around synthesizing the bulk gCN, opting for unconventional exfoliation strategies, and making composites with metal and non-metal components. Therefore, the thesis is divided into **five chapters**:

The **first chapter** comprehensively introduces gCN, covering its brief history, structural features, advantages, various preparation methods, and limitations regarding electrocatalytic activity. Furthermore, the first chapter addresses the various methods of catalyst immobilization on conductive electrodes, along with their associated drawbacks. At the conclusion of the first chapter, the rationale for selecting the electrodeposition protocol for gCN immobilization on the electrode was discussed, along with a detailed explanation of the methods employed for electrodeposition and catalytic activity testing, specifically Cyclic Voltammetry and Square Wave Voltammetry.

The **second chapter** of the thesis consists of two distinct sections. The first part addresses the synthesis, characterization, and electrodeposition of bulk and ball-milled gCN. This chapter features the step-by-step mechanistic understanding of the electrodeposition and the protocol optimization in terms of scan rate, number of scans, and material. The electrodeposition was confirmed through in-depth spectroscopic, microscopic, and electrochemical tools. The chapter further presents the incorporation of MnO_2 to achieve a synergistic enhancement of electrocatalytic activity. The electrodeposition of gCN.MnO_2 was carried out using two strategies. Firstly, gCN.Mn_xO_y was synthesized and then the electrodeposition was carried out. In the second approach, the in-situ formation and electrodeposition of gCN.MnO_2 on more viable, screen-printed electrode was targeted. The electrocatalytic activity of the modified surfaces were characterized by comparing the enhancement in the peak current (sensitivity) and the shift in the peak potential for an ideal redox probe $K_3[Fe(CN)_6]$ and for the dopamine electro-oxidation, a widely recognized neurotransmitter.

In the **third chapter**, the gCN.CuO composite was synthesized and characterized. The CuO is specifically selected to enhance the electrochemical performance of gCN by means of the interaction between the π-π electron cloud and with high spin density N of polymeric gCN structure and the vacant d orbitals of Cu(II). The synthesized composite was electrodeposited on a glassy carbon electrode and subsequently characterized. Following the electrodeposition, the gCN.CuO composite exhibited a distinctly altered morphology characterized by nano-flakes. The electrochemical performance of the modified and unmodified surface was then compared for the investigation of the electro-oxidation of an ideal redox probe $K_3[Fe(CN)_6]$. Lately, due to the complexing ability of CuO with Riboflavin, the modified electrode was used for the electroanalysis of Riboflavin.

After modifying the gCN with metals, the **fourth chapter** addresses tuning the electrocatalytic activity of gCN with phosphorus doping. gCN with three distinct P-doping percentages were synthesized and thoroughly characterized. The electrocatalytic activity of the P-doped gCN modified surface was studied using an ideal redox probe $K_3[Fe(CN)_6]$. The best-performing material was then employed to study the interfacial electrochemistry of tryptophan for qualitative and quantitative analysis.

The **fifth chapter** of this thesis summarizes the research comparing the electrocatalytic activity of all the mentioned composites, draws conclusions, and discusses future perspectives related to this work.