Chapter 1:
Introduction

1.1 Energy Crisis: Challenges and Solutions

In recent years, the world has witnessed a surge in energy demands driven by technological advancements and rapid population growth. This, coupled with decline of traditional fossil fuel such as petrol, diesel, etc., has necessitated the search for alternative fuels, particularly in the transportation sector.[1] One promising candidate that has garnered significant attention is hydrogen fuel which possesses several desirable characteristics making it an attractive alternative to conventional fuels. Firstly, it is abundantly available, with sources ranging from water to biomass and renewable energy resources.[2] Unlike fossil fuels, hydrogen is considered eco-friendly since it produces only water and heat when used as a fuel, avoiding harmful emissions such as greenhouse gases.[2] Additionally, hydrogen boasts a high energy density, meaning it can store and deliver more energy per unit of mass in comparison to alternative fuels. However, the practical use of hydrogen fuel faces two main challenges: efficient and cost-effective production and compact storage.[3,4] This thesis addresses these challenges by designing advanced two-dimensional (2D) nanomaterials to produce hydrogen through water splitting, as well as exploring efficient methods for hydrogen storage.[3,4]

1.2 Water Splitting Method for Hydrogen Production

In recent times, water splitting has gained considerable attention and emerged as a promising approach for hydrogen production.[5] This process encompasses two distinct half reactions that occur simultaneously: (I) hydrogen evolution reaction (HER) taking place at the cathode and (II) oxygen evolution reaction (OER) occurring at the anode.[5] By means of these reactions, water molecules undergo transformation, resulting in the generation of hydrogen gas

and oxygen gas, respectively. The occurring of HER at cathode leads to the reduction of water molecules and the subsequent production of hydrogen gas. During this reduction process, electrons are transferred from an external power source to the water molecules, inducing a sequence of chemical transformations. As a result, hydrogen ions (H^+) from the water molecules acquire these electrons and subsequently combine to form hydrogen gas.[3] The HER plays a pivotal role in the process of water splitting as it directly facilitates the generation of renewable and clean hydrogen fuel, which holds immense potential as a sustainable energy source. Enhancing the performance of water splitting systems relies heavily on the development and optimization of efficient catalysts. Catalysts are substances that aid in reactions by lowering the necessary activation energy and promoting faster rates of reaction.[6] In the context of water splitting, catalysts are employed at both the cathode and the anode to enhance the efficiency of the HER and OER, respectively. Numerous catalysts have been studied for the process of water splitting, which include noble metals like platinum and iridium oxide, as well as alternative catalysts based on non-precious metals such as transition metal oxides and/or sulfides.[7,8] These catalysts have the potential to greatly enhance the kinetics of the HER and OER, thereby increasing the efficiency and cost-effectiveness of the water splitting process. However, challenges remain in terms of finding catalysts that are abundant, inexpensive and stable under the harsh operating conditions of water splitting systems. In addition to catalyst development, research efforts have also focused on exploring different methods for water splitting such as photoelectrochemical and electrolysis-based approaches.[9,10] The photoelectrochemical water splitting utilizes light energy to drive the reactions, employing semiconducting materials that can absorb photons and generate electron-hole pairs. These charge carriers then participate in the HER at the respective electrodes, resulting in the separation of water into hydrogen.[9,10] In contrast, electrolysis-based water splitting depends on the utilization of an external electric potential to induce and drive the reactions.[9,10] Water

splitting holds tremendous promise for sustainable hydrogen production, as it utilizes water as the raw material and yields hydrogen gas, a clean and high-energy-density fuel. The hydrogen produced through water splitting can be utilized in fuel cells for power generation, as a transportation fuel and in various industrial processes.[9,10]

1.3 Hydrogen Storage: Challenges, Methods and Advancements

Designing advance materials for high-capacity, recyclable hydrogen storage devices present a significant challenge due to limitations with current hydrogen storage technologies.[2] In the context of fuel, a typical car would burn 24 kg of petrol to cover a distance of 400 km. However, the car can travel the same distance only using 4 kg of hydrogen.[11,12] Hydrogen is in a gaseous form and to store 4 kg of hydrogen at room temperature and pressure, approximately 45 m^3 of volume is required.[11,12] This volume would be greater than the size of the car itself, making the storage of hydrogen in a small and lightweight material is one of the biggest challenges faced by onboard automobile applications.[11,12] Currently, physical or chemical methods are used for hydrogen storage. One physical storage method is high-pressure storage, where hydrogen is stored at high pressure in cylinders or tanks. Nevertheless, this approach suffers from significant drawbacks, including the bulky and weighty nature of the tank, as well as the potential hazard of tank rupture caused by the extremely high pressure of hydrogen.[11-13] Another physical hydrogen storage method is liquid state storage, where hydrogen is stored in its liquid phase below 20 K temperature with liquid helium as a coolant. The expensive process of liquefying hydrogen gas stands out as a significant drawback of this method.[11-13] Hence, the safety and economic considerations of hydrogen storage have attracted the scientific and research community to explore solid-state storage techniques, specifically involving the adsorption of hydrogen onto abundant terrestrial materials.[14] Solid-state storage offers a promising solution as a highly desirable and efficient alternative, given that the storage material can effectively absorb significant quantity of hydrogen and release it effortlessly, while

maintaining its structural integrity.[14] The “U.S. Department of Energy (DoE)” has set guidelines for efficient hydrogen storage devices. According to these guidelines, a system should be capable of storing hydrogen at a minimum of 6.5 wt.% by total weight, while the binding energy of hydrogen molecules must fall within the range of 0.2 - 0.8 eV.[15]

1.4 Two-dimensional (2D) Materials

1.4.1 Graphene: Pioneering the Era of 2D Materials

Graphene, often hailed as a "wonder material," is an extraordinary substance that has revolutionized the field of materials science. Its story begins with the curiosity of two scientists (Andre Geim and Konstantin Novoselov), who made an extraordinary discovery in 2004.[16] These scientists were investigating the properties of carbon, a well-known and abundant element. Their curiosity was specifically piqued by graphite, a substance identical to the one commonly used in pencil leads. Graphite is composed of carbon atoms ordered in a hexagonal lattice structure, forming stacked layers. In an experiment, the two scientists used a simple method to isolate single layer of graphite from bulk material. They took a piece of sticky tape and repeatedly peeled it off the graphite, hoping to obtain thinner layers. Surprisingly, they managed to isolate a layer that was incredibly thin-only one atom thick.[17] This incredibly thin layer was graphene, a 2D material consisting of a single atomic-thick layer of carbon atoms organized in a hexagonal lattice pattern. The groundbreaking finding of graphene by Geim and Novoselov led to their Nobel Prize win for physics in 2010.[18] This discovery marked the dawn of a new direction in materials exploration, showcasing immense possibilities for various applications.[19] Graphene possesses remarkable properties that make it truly exceptional. It is the thinnest material ever discovered, but also one of the strongest, around 200 times stronger than steel.[20] Graphene exhibits unparalleled electrical conductivity surpassing that of a few well-known compounds and it also possesses exceptional thermal conductivity. Additionally, it is flexible, transparent and impermeable to most gases and liquids.[20] The unique combination

of these properties sparked immense interest and excitement within the scientific community and various industries. Researchers around the world have started exploring the potential of graphene in various fields, including electronics, energy storage, biomedical devices and many more.[21,22] In the electronics industry, graphene has the potential to revolutionize computing and communication devices. Its high electron mobility makes it an ideal material for ultrafast transistors, leading to faster and more efficient electronic devices.[22] Superior electrical conductivity of the graphene also makes it compatible for flexible displays and touchscreens.[23] In the energy sector, graphene-based materials have been explored for high-performance batteries, supercapacitors and solar cells. Graphene's ability to store and conduct energy efficiently could lead to longer-lasting and faster-charging batteries, while its transparency and flexibility make it an excellent candidate for next-generation solar panels.[21,24] Graphene's applications extend beyond electronics and energy. The biocompatibility and substantial surface-to-volume ratio of graphene have garnered significant attention in the advancement of biosensors and drug delivery systems in the field of medical diagnostics.[24,25] Despite its incredible potential, graphene still faces challenges in large-scale production and integration into existing manufacturing processes.[26] Researchers are actively working to overcome these hurdles and unlock the full potential of graphene-based technologies. In conclusion, discovery of graphene by Andre Geim and Konstantin Novoselov marked a significant milestone in materials science.[21-26] Its exceptional properties have captivated scientists and engineers worldwide, and ongoing research continues to uncover new and exciting applications for this remarkable material. [21-26] Graphene's story is still unfolding and its impact on various industries and our everyday lives holds great promise for the future.[21-26]

1.4.2 Beyond Graphene: Advancements in 2D Materials

Since the discovery of graphene, significant progress has been made in the field of 2D materials. Researchers have actively explored and synthesized a wide range of 2D materials,

each with diverse properties and applications.[27] Here are some notable advances in 2D materials following graphene. One such material is 2D hexagonal boron nitride (h-BN), which shares a similar structure to graphene and shows great potential for various applications, including photonics, fuel cells and serving as a substrate for 2D heterostructures.[28] While both graphene and 2D h-BN possess an isostructural nature, graphene exhibits conductivity, whereas 2D h-BN acts as a wide-gap insulator.[29] Furthermore, graphyne, akin to graphene, is a 2D carbon allotrope that shares a similar structural pattern.[30] It can be visualized as a lattice comprising interconnected benzene rings joined by acetylene bonds. Graphyne exhibits a mixed hybridization denoted as sp^n, with $1 < n < 2$, depending on the presence of acetylene groups.[30] This stands in contrast to the pure sp^2 hybridization of graphene and the pure sp^3 hybridization of diamond.[31] Using first-principles calculation incorporating phonon dispersion curves and ab initio molecular dynamics (AIMD) simulation, it has been demonstrated that graphyne and its boron nitride counterparts exhibit stability.[32] In a similar context, graphdiyne, which is a variant of graphyne containing diacetylene groups, was successfully created on copper substrates.[33] These materials possess unique properties that render them potentially valuable in a wide range of applications, including electronics, energy storage, catalysis, sensors, gas storage and separation.[31] After the discovery of carbon-based graphene, mono-elemental 2D materials also gained significant attention in the field of materials science. In this regard, borophene to bismuthine monolayers were theoretically predicted and experimentally realized.[34] These mono-elemental 2D materials have also been proven for various applications in different fields.[34]

In the direction of the 2D materials, transition metal dichalcogenides (TMDs)[35], such as molybdenum disulfide (MoS_2) and tungsten diselenide (WSe_2), are well-known examples of 2D materials. These materials have attracted significant attention due to their unique properties and potential applications in various fields. TMDs offer a wide range of electronic, optical and

mechanical properties, making them suitable for applications in flexible electronics, optoelectronics and sensors.[35] MXenes are a family of 2D materials that exhibit high electrical conductivity and good mechanical strength. They are derived from layered ceramics known as MAX phases by selectively etching the "A" element. MXenes show promise for various applications, including energy storage devices (batteries and supercapacitors), electromagnetic interference shielding and catalysis.[36] In recent times, there has been a significant surge of interest in binary 2D materials belonging to groups III–VI, IV–VI, V–VI, III–V, IV–V, and V–V, making them a prominent topic of research.[37-41] Graphitic carbon nitride[37], commonly referred to as g-C_3N_4, has garnered considerable interest in recent times as a 2D material. Its structure comprises carbon and nitrogen atoms arranged in a lattice reminiscent of honeycombs, akin to graphene. However, in contrast to graphene, g-C_3N_4 acts as a semiconductor with a moderate bandgap. Additionally, g-C_3N_4 has been explored as a material for energy storage and conversion devices such as lithium-ion batteries, supercapacitors and fuel cells. Its unique structure and electronic properties contribute to its potential in these fields by facilitating charge transport and storage.[37] Furthermore, one of the popular experimentally discovered 2D materials from group III-V is gallium nitride (GaN), which is used for producing light-emitting diodes.[38] 2D materials indium selenide (InSe) is composed of elements from group III–VI which is layered semiconductor material that exhibits strong light-matter interactions and has potential applications in photovoltaics, transistors and photodetectors.[39]

Motivated by the above experimentally synthesized 2D materials, the theoretical prediction and experimental validation have become hot topics for research. In a recent study, Qingxing et al.[40] investigated group IV-V-VI monolayers such as P_2SiS, As_2SiS, As_2GeSe, Sb_2GeSe, Sb_2SnTe and Bi_2SnTe. Computational techniques were used to assess stability, revealing that all monolayers exhibit semiconductor properties. This expands the range of group V semiconductors and paves the way for discovering new semiconductors with improved

characteristics. These advancements have significant potential for optoelectronic and electronic applications in 2D materials.[40] Recently, Burak et al.[41] successfully predicted monolayers based on group IV and V elements with A_2B_2 chemical formula (where A = C, Si, Ge, Sn and Pb, and B = N, P, As, Sb and Bi) using first-principles calculations and discovered two dynamically stable structural symmetries in the form of hexagonal lattices, namely $P\bar{6}m2$ and $P\bar{3}m2$, which were respectively named the α- and β-phases.[41] Moreover, the prediction of unexplored materials is vital for the advancement of material science. It provides a powerful tool for identifying materials with desired properties and enables researchers to explore their potential applications. By integrating computational predictions with experimental techniques, scientists can uncover new materials and drive innovation in various industries.

1.4.3 2D Materials for Water Splitting

In recent times, single-atom catalysts (SACs) have emerged as a promising solution for advancing clean and sustainable energy technologies.[42] They offer benefits like single active sites, strong metal-support interactions and impressive catalytic activity for HER.[42] Additionally, 2D materials show promise as supports for SACs in HER. Pristine graphene doesn't interact well with hydrogen atom, so it's not suitable for HER activity.[43] Researchers conducted density functional theory (DFT) simulations and experiments to study SACs of transition metals on N doped graphene. During their research, it was discovered that the Rh, Cr, Co, V and Fe exhibited favourable activity for the HER, with Co-SAC demonstrating the lowest Gibbs free energy (ΔG_H) (-0.20 to 0.30 eV). As a result, Co-SAC proved to be an exceptionally efficient catalyst for HER.[44] Dušan et al.[45] employed DFT study to discover effective SACs featuring transition metals inserted into the monovacancies of h-BN and graphene. They examined a total of 27 SACs, among which several candidates exhibiting HER activity comparable to that of Pt surfaces were identified.[45] DFT simulations were performed to study HER activity on graphyne supported SACs. Among the tested transition metals (Cu,

Fe, Zn, Ni and Co), Ni SAC displayed the excellent HER activity (0.08 eV).[46] Xue et al. investigated the use of graphdiyne as a support for SACs in HER activity, focusing on Ni and Fe as active metals. DFT calculations indicated that Ni and Fe SACs anchored on graphdiyne exhibited favourable HER activity.[47] Sun and colleagues conducted a study that revealed the poor catalytic activity of pristine silicene for HER.[48] However, when metal atoms (Mn, Ni, Ti, Fe, Be, V, Co and Cr) were anchored over surface of silicene, it exhibited good catalytic activity for HER.[48] The calculated ΔG_H values ranged from -0.09 to 0.18 eV, which is in close proximity to the ideal value for the HER activity. Additionally, the research conducted by Zhu et al. emphasizes the enhanced photocatalytic HER activity performance achieved by loading g-C_3N_4 with Pt metal, resulting in the maximum utilization efficiency of Pt atoms.[49] Apart from the mentioned materials, various 2D supports (such as C_2N, C_9N_4, etc.) are currently being intensively studied for SACs.[50,51]

In addition to SACs, non-transition metal doping (e.g., B, N, C, etc.) and defects engineering are also highly effective methods for enhancing the HER activity of 2D materials.[52,53] In this context, the HER activity of the graphene monolayer was found to increase with the B, S and N dopants.[54,55] Similarly, pristine g-C_3N_4 is also not suitable for HER activity, but the dopants S, Na and O enhance the HER activity as compared to its pristine counterpart.[56,57] Furthermore, Deobrat et al. conducted a study highlighting the significant impact of C, S and P doping on the HER activity of the Si_2BN monolayer, effectively enhancing it.[58] In a study conducted by Wan et al., it was demonstrated that the porous boron nitride (p-BN) possesses insulating properties with an exceptionally wide band gap, akin to a h-BN monolayer. Consequently, p-BN alone is deemed unsuitable for catalytic activities. However, when C is introduced as a dopant in p-BN, the band gap undergoes tuning, rendering it a viable metal-free photocatalyst capable of facilitating overall water splitting when exposed to visible light irradiation.[59] In a previous study, Yixin and colleagues provided evidence that the

introduction of various defects can alter the electronic characteristics of MoS_2, resulting in a substantial improvement in its HER activity.[60]

In summary, the exploration of predicted and experimentally synthesized materials as supports for SACs continues to be an immensely intriguing research area with numerous applications. Additionally, researchers have shown significant interest in investigating the modulation of structural and electronic properties through substitutional doping and the introduction of defects, aiming to enhance the HER activity of the materials under consideration.

1.4.4 2D Materials for Hydrogen Storage

Hydrogen storage is a crucial factor in the effective utilization of hydrogen as a clean energy carrier. Recently, the utilization of 2D materials for solid-state hydrogen storage has garnered considerable interest owing to their distinctive properties such as high surface area, tunable electronic structure and exceptional mechanical strength. These properties make them highly promising for hydrogen storage applications.[14] Specifically, the substantial surface area of graphene and its capability to adsorb hydrogen molecules on its surface make it an appealing candidate. Nonetheless, the binding energy between hydrogen and graphene is relatively weak, necessitating further advancements to enhance the storage capacity.[61-63] Enhancing hydrogen storage capacity in graphene has proven challenging. Researchers have tried various approaches such as decoration of metal atoms (alkali metals, alkaline earth metals and transition metals), creation of defects and strain to enhance storage capacity of graphene.[61-63] In this regards, Yali et al. successfully improved the storage properties of hydrogen by adding Ti metal into graphene, resulting in significant improvements in storage parameters.[61] Investigations have also explored the potential of Y doped graphene as a reversible and efficient material for hydrogen storage, demonstrating a capacity of 6.17 wt%.[62] Miao et al. conducted a study on strain-engineered graphene decorated with metal atoms for hydrogen storage and

found that applying a 10% strain to a graphene monolayer increased the adsorption strength of Li and Ti atoms by approximately 75% and 71% respectively. This modification induced by strain enabled hydrogen storage densities that surpassed the limits set by the DoE.[63] Furthermore, Metal-organic frameworks (MOFs) and TMDs are promising 2D materials for hydrogen storage. MOFs consist of metal ions and organic ligands, offering a porous structure with high surface area and adjustable properties. Various MOFs have been synthesized and modified to optimize hydrogen interaction and increasing storage capacity.[64] TMDs such as MoS_2 and WS_2 have layered structures with strong van der Waals interactions between layers.[65] These materials offer adsorption sites for hydrogen storage. However, by introducing defects or functionalizing the surface, the hydrogen storage capacity of TMDs can be further improved.[65] Extensive research has focused on h-BN monolayer, an allotrope of graphene, for its potential applications. Initially, it was found that pure h-BN monolayer is not ideal for hydrogen storage. However, scientists found that by introducing Ti atoms into C doped h-BN, the hydrogen storage capacity can be greatly enhanced.[66] Jing and colleagues explored the properties of expanded hexagonal boron nitride (eh-BN) as a hydrogen storage medium. They determined that the maximum hydrogen gravimetric density in eh-BN was 2.96 wt% at 243K and 10MPa, which falls below the standard set by the DoE.[67] Similarly, various techniques, such as Ti doping into divacancy h-BN, hydrogenation of h-BN with Li and the decoration of Ni on defective h-BN, have demonstrated substantial enhancements in hydrogen storage characteristics compared to the bare monolayer.[68-70] In addition, Zhang and colleagues achieved an increased hydrogen storage capacity of 7.5 wt% by introducing Li doping into the porous boron nitride sheet.[71] Apart from these, various pristine and functionalized monolayers such as psi-graphene, Si_2BN, etc., are widely studied for hydrogen storage applications.[72,73]

In today's fast-paced and ever-evolving world, it is absolutely crucial to stay informed about the latest breakthroughs in predicted and experimentally synthesized materials. This

knowledge holds immense significance as it allows us to evaluate the potential applications of these materials and tap into their vast possibilities. By actively keeping up with the developments in this field, we gain the ability to identify and explore new avenues for utilizing these materials, propelling innovation forward and fostering collaborative efforts across different disciplines. Ultimately, this collective knowledge empowers scientists to drive transformative advancements across a wide range of industries, opening up a world of endless opportunities and pushing the boundaries of what is possible.

1.5 Materials and Methodology for Research Work

Recently, Burak et al. successfully predicted dynamically stable hexagonal α-SiX (X = N, P, As, Sb and Bi) monolayers based on group IV and V elements with the A_2B_2 chemical formula using first-principles calculations.[41] Furthermore, Demirci et al. predicted an orthorhombic diboron dinitride (o-B_2N_2) monolayer with remarkable thermal and dynamic stability, possessing an orthorhombic lattice structure and demonstrating semiconductor properties characterized by a band gap of 0.64 eV.[74] Very recent, holey graphyne (HGY), a 2D graphyne analogous structure, was recently synthesized experimentally from Castroe-Stephens coupling reaction from 1,3,5-tribromo-2,4,6-triethynyl benzene.[75] HGY features uniform pores with six, eight and twenty-four vertex rings surrounded by zero, two and six acetylene linkages (sp-sp bonds). The carbon atoms of the benzene ring connect to the nearest octagon's carbon atoms, resulting in 0.50 sp to sp^2 carbon ratio. Moreover, the aforementioned predicted/synthesized materials have not yet been explored as catalysts for the HER and efficient hydrogen storage medium.

In the current thesis, we examined the HER activity of several unexplored materials, including α-SiX (X = N, P, As, Sb, and Bi)[76], holey graphyne[77] and o-B_2N_2 monolayers[78], using dispersion-corrected DFT calculations performed with the Quantum Espresso code. We studied the α-SiX and holey graphyne monolayers to support SACs in order to enhance the HER

activity performance. Additionally, we examined the o-B_2N_2 monolayer as an HER catalyst under pristine, defected and C doped conditions. As a result, functionalizing these monolayers under the mentioned conditions leads to desirable outcomes. Furthermore, we have also assessed the thermal stability of the catalysts that exhibited good HER activity from AIMD simulations at room temperature using Vienna Ab Initio Simulation Package (VASP) software. These simulations have demonstrated favourable outcomes in terms of its stability under thermal conditions.

In the current thesis, our research delved into the hydrogen storage capabilities of the recently predicated o-B_2N_2 monolayer. We undertook an extensive analysis of its hydrogen storage potential across various scenarios, encompassing its pristine state, instances of defects, and under different metals (such as Na, K, Be, Mg, Ca, Sc, Ti, Y and Zr) decoration. Notably, our investigation revealed that the o-B_2N_2 monolayer decoration with Ti metals exhibited remarkably promising outcomes as a hydrogen storage medium, satisfying all the outlined criteria stipulated by DoE. In addition, our AIMD findings have highlighted the robust thermal stability of the Ti decorated o-B_2N_2 monolayer, even when exposed to desorption temperatures relevant to hydrogen release. This underscores the material's effectiveness for hydrogen storage and its practical application.

1.6 Structure of the Present Thesis

The rest of this thesis are organized as follows:

Chapter 2 provides a comprehensive overview of the computational methodology employed in this study. The chapter explores the theoretical foundations of DFT, which serves as the fundamental framework for calculating various properties of 2D materials. Key concepts including the Born-Oppenheimer approximation, Hartree approximation, Hartree-Fock approximation and density-based methods are discussed in detail. Furthermore, chapter 2

examines the importance of the Kohn-Sham equation in DFT applications. This chapter also explains the use of plane waves to represent electron wave functions and discusses the density calculation techniques required for addressing valence and core electrons. Special emphasis is given to the implementation of these methods in the Quantum Espresso simulation code.[79] Furthermore, AIMD method used to investigate the thermal stability of materials at specific temperature is also briefly explained. This approach involves simulating the dynamics of atoms based on first principles to determine how the material behaves under varying thermal conditions.

Chapter 3 presents our work on transition metals supported by α-SiX (X = N, P, As, Sb, Bi) and holey graphyne (HGY) monolayers as SACs to enhance the HER activity.[76,77] We systematically discuss the structural and electronic properties of both pristine and transition metals anchored monolayers as SACs using DFT simulations. Additionally, we analyze the effects of transition metals on the supports by studying the partial density of states (PDOS), electronic band structure, charge transfer and other relevant factors. To evaluate the suitability of the HER activity, we examine the ΔG_H of the hydrogen adsorbed systems and compare them with previous reports. Furthermore, we discuss the room temperature stability by employing AIMD calculations for good HER catalysts derived from our investigation.

Chapter 4 discusses the structural and electronic properties, HER activity and stability of the pristine, vacancy and C doped 2D o-B_2N_2.[78] This chapter includes a comparative analysis of the structural and electronic properties before and after introducing defects and C doping. For the HER analysis, we present the obtained ΔG_H values of the hydrogen adsorbed pristine, defected and C doped monolayers and compare them with previous reports.

Chapter 5 focuses on studying the hydrogen storage application of the newly predicted o-B_2N_2 monolayer. We discuss the suitability of defected o-B_2N_2 monolayers, as well as o-

B_2N_2 monolayers decorated with metals (Na, K, Be, Mg, Ca, Sc, Ti, Y, and Zr), for hydrogen storage. Our discussion in this chapter involves the calculation of various properties such as average adsorption energy of the hydrogen, hydrogen gravimetric density, desorption temperature and other electronic parameters using dispersion-corrected DFT simulations. We also validate the room temperature stability of the best case through AIMD simulations.

Chapter 6 serves as a conclusive overview, encapsulating the pivotal findings and prospective applications derived from this thesis. It also delves into the prospects for future research in the domain of water splitting and hydrogen storage, focusing on the utilization of 2D materials.

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