DFT-Guided Design and Fabrication of Carbon-Nitride-Based Materials for Energy Storage Devices: A Review

David Adekoya¹, Shangshu Qian¹, Xingxing Gu¹, William Wen¹, Dongsheng Li², Jianmin Ma³, Shanqing Zhang¹

HIGHLIGHTS

• Comprehensive summary of crystalline structures and morphologies of carbon nitride-based materials (CNBMs).
• Density functional theory computation for the design of functional CNBMs for rechargeable battery applications.
• The experimental synthesis strategies of CNBMs for rechargeable battery application.

ABSTRACT Carbon nitrides (including CN, C₂N, C₃N, C₃N₂, C₄N, and C₅N) are a unique family of nitrogen-rich carbon materials with multiple beneficial properties in crystalline structures, morphologies, and electronic configurations. In this review, we provide a comprehensive review on these materials properties, theoretical advantages, the synthesis and modification strategies of different carbon nitride-based materials (CNBMs) and their application in existing and emerging rechargeable battery systems, such as lithium-ion batteries, sodium and potassium-ion batteries, lithium sulfur batteries, lithium oxygen batteries, lithium metal batteries, zinc-ion batteries, and solid-state batteries. The central theme of this review is to apply the theoretical and computational design to guide the experimental synthesis of CNBMs for energy storage, i.e., facilitate the application of first-principle studies and density functional theory for electrode material design, synthesis, and characterization of different CNBMs for the aforementioned rechargeable batteries. At last, we conclude with the challenges, and prospects of CNBMs, and propose future perspectives and strategies for further advancement of CNBMs for rechargeable batteries.

KEYWORDS Carbon nitrides; Metal-ion batteries; Density functional theory; g-C₃N₄; Anode
1 Introduction

Rechargeable metal ion batteries (MIBs) are one of the most reliable portable energy storage devices today because of their high power density, exceptional energy capacity, high cycling stability, and low self-discharge [1, 2]. Lithium-ion batteries (LIBs) remain the most developed and commercially viable alternative among all rechargeable batteries, and graphite is widely accepted as a preferred negative electrode (anode) material for LIBs [3]. Graphite is affordable, and it does not suffer significant volumetric changes (compared to other metal-based electrodes); it operates at a very low voltage close to that of lithium metal (~0.1 V) and exhibits moderately stable cycle life with a theoretical capacity of 372 mAh g⁻¹ (LiC₆). Despite these positive characteristics, the performance of graphite is still very limited, and one of the effective ways to modulate its properties and electrochemical performance is through nitrogen doping [4]. Precisely, the work of Liu et al. showed that the N-doping of graphene results in improved conductivity and charge transfer, which boosts its performance and cyclability [5].

Carbon nitrides are a family of nitrogen-rich graphite analogues which contain a high nitrogen content and porous defect sites for effective charge transfer in energy storage devices [6, 7]. However, carbon nitrides are limited by poor electrical conductivity, chemical inertness, and ineffective intercalation/deintercalation process [8]. Due to these issues, several research studies have focused on the design of unique carbon-nitride-based materials (CNBMs), including pure carbon nitrides, doped carbon nitrides (DCNs) as well as carbon-nitride-based composites (CNBCs) [9]. Most of these reports have focused on g-C₃N₄ because it is easy to synthesize, low cost, environmentally safe, and it has a theoretical capacity of 524 mAh g⁻¹ (Li₂C₃N₄). In contrast, though other carbon nitride structures (such as CN, C₂N, C₃N, C₄N, and C₅N) which have been studied through density functional theory (DFT) calculations exhibit exceptional structural and electronic properties which demonstrates their capabilities as promising MIB electrode materials, they have attracted less attention.

On the other hand, DFT has been adopted more and more in studying CNBMs and their electronic properties [10]. In fact, DFT could provide effective guidance for the synthesis of electrode materials and/or interpretation of the structure–performance relationship in energy storage devices, including LIBs, sodium- and potassium-ion batteries (SIBs and PIBs), lithium-sulfur batteries (Li–S), lithium-oxygen (Li-O₂) batteries, lithium metal batteries (LMBs), zinc air batteries (ZABs), and solid-state batteries (SSBs). This review aims to comprehensively discuss the relationships between the structural and electronic properties and the MIB performance of pure carbon nitrides, doped carbon nitrides, and CNBCs, summarizes the theoretical computation for the design of functional CNBMs for different rechargeable MIB applications, and generalizes the synthesis strategies of pure carbon nitrides and CNBCs for rechargeable metal ion battery application (Fig. 1). At the end of this work, we also offer a perspective on the existing challenges of carbon nitrides for energy storage devices and relevant resolving strategies.

2 CNBMs

To study the carbon nitrides, we must begin by exploring their crystalline structure, structural and electronic properties, and functionality. Carbon nitrides are a family of nitrogen-rich carbon materials, and they have different crystalline structures and molecular configurations. Generally, there are seven types of nitrogen species (Fig. 2), with at least two most common species of nitrogen (the graphitic-N and pyridinic-N). The nature of the nitrogen and its percentage concentration has been proven to impact the electronic configuration and characteristic of the carbon nitride for different applications. The classification of carbon nitrides is often based on the carbon to nitrogen content (i.e., the C/N ratio) which is often associated with the degree of surface defects in their structure [11]. The carbon nitrides discussed in this review are categorized based on the dominant N species in their crystalline structure.

2.1 Pure Carbon Nitrides

Based on their generic family molecular formula of CₓNᵧ, the carbon nitride family includes CN (C₂N₂, C₃N₃, C₄N₄), C₂N, C₃N, C₃N₂, C₄N₄, C₃N₅, C₃N₆, C₃N₇, C₄N, C₄N₃, C₅N, C₆N₆, C₆N₈, C₉N₄, C₉N₇, C₁₀N₃, C₁₀N₉, and C₁₄N₁₂ [11, 13]. All these carbon nitride materials possess some similarity with graphene in that they are all two-dimensional (2D) materials with sp²/sp³ hybridized conjugated C atoms.
but they all exhibit different structural frameworks and C/N ratio. In this section of the review, we summarize the crystalline structure, surface functionalities, and electronic properties of different carbon nitrides, including pyridinic nitrogen-based carbon nitrides (CN, C$_2$N, and C$_3$N$_4$) and graphitic nitrogen-based carbon nitrides (C$_3$N, C$_4$N, and C$_5$N) (Fig. 3).

**2.1.1 Pyridinic Nitrogen-Based Carbon Nitrides (CN, C$_2$N, C$_3$N$_4$)**

Pyridinic-N is one of the edge site nitrogen species in an N-doped carbon material or carbon nitride. It is bonded to two carbon atoms and supplies one p electron to the $\pi-\pi$ conjugated system of carbon [12]. Due to the co-ordination of pyridinic-N to 2 carbon atoms, it donates 4 electrons to the $sp^2$ of each carbon atom along with a lone pair. Because of its superior electronegativity, it is able to pull $\pi$-electrons from the conjugated system of carbon, thereby making it negatively charged N-atom. When the $\pi$-conjugated system of carbon is broken (the aromaticity), a non-bonding $p_z$ orbital is formed, and this orbital can also be observed around defected carbon materials. Carbon materials that contain pyridinic-N can function as Lewis base because the
pyridinic-N exhibits a negative charge on surrounding carbon atom, which makes them positively charged with a lone pair of electrons as a result of the non-bonding $p_z$ orbital [12]. The pyridinic-N-based carbon nitrides in this category include CN, $C_2N$, and $C_3N_4$.

2.1.1.1 CN CN exists in different compositions such as $C_2N_2$, $C_3N_3$, and $C_4N_4$, and the structure of these carbon nitride materials possesses round and uniform pores, six pyridinic nitrogens in each unit cell and a bandgap of $\sim 1.5\text{--}1.6$ eV (Fig. 4a, b) [15]. Most of the carbon nitrides contain both graphitic and pyridinic nitrogen except for CN. Due to its rich pyridinic nitrogen structure and absence of graphitic nitrogen, CN exhibits high structural stability, better conductivity and does not experience loss of crystallinity when it interacts with alkali metal ions like $Li^+$. 

2.1.1.2 $C_2N$ The structure of $C_2N$ exhibits $sp^2$ hybridization, and it is filled with uniformly sized holes and one large hole at the center, which is due to the six-member nitrogen-containing ring (Fig. 4c). Like the structure of graphene, it possesses several benzene rings that are bridged by pyrazine rings, which have two nitrogen atoms opposite to each other [16]. Also, unlike graphene, which has a fully conjugated $\pi$-electron structure, the presence of nitrogen in the structure of $C_2N$ makes the $\pi$-electronic structure of its benzene rings isolated. Therefore, it has a flat band and exists as a semiconductor with a bandgap of 1.96 eV (Fig. 4d). Due to its wider structure and the abundance of several benzene and pyrazine rings, it is expected that it will possess several active sites for alkali metal ion storage. Moreover, the defect sites and higher nitrogen population of $C_2N$ suggest that it will exhibit superior conductivity to boron nitride (BN) [17].

2.1.1.3 $C_3N_4$ $C_3N_4$ carbon nitride is the most reported member of the carbon nitride family. It is composed of consistently repeated tri-s-triazine units having planar $sp^2$-hybridized conjugation structures held together by van de Waals forces. It often occurs as bulk $C_3N_4$, which has a relatively low indirect bandgap of $\sim 2.7$ eV and an interplanar distance of 0.324 nm and can be functionalized to obtain other architectures such as the monolayer 2D sheet (Fig. 4e) [18]. This indirect bandgap of $C_3N_4$ highly has spurred massive interest in its application for

![Fig. 3 Geometric structures of the pyridinic and graphitic-N-based carbon nitrides reported for rechargeable batteries and their surface functionalities. The brown spheres represent carbon atoms, and the light blue one represents nitrogen atoms in the 2D carbon nitrides structures. (Color figure online)](image-url)
several applications such as photocatalysis, oxygen evolution, reduction reactions, sensing devices, etc. However, the semiconductor property of bulk C\textsubscript{3}N\textsubscript{4} limits the electronic conductivity for application in electronic devices [19]. Under buckling, the indirect bandgap of bulk C\textsubscript{3}N\textsubscript{4} can be tuned into a direct bandgap material with superior electronic conductivity (Fig. 4f) [20]. This ability to easily modify the electronic and structural properties of C\textsubscript{3}N\textsubscript{4} makes it one of the most preferred and widely applied carbon nitride structures [21].
2.1.2 Graphitic Nitrogen-Based Carbon Nitrides (C$_3$N, C$_4$N, and C$_5$N)

Graphitic nitrogen is one of the most widely known forms of nitrogen. It has a nitrogen bonded to three carbon atoms, and it is the nitrogen-doped into the basal graphitic carbon plane of the material. Due to its superior negativity in comparison with carbon, it is possible for graphitic nitrogen to induce a positive charge on adjacent carbon, thereby making it a readily active site for attraction to negatively charged species [22]. This makes it a commonly desired form of nitrogen doping, just like pyridinic nitrogen. The graphitic-N-based carbon nitrides, which we shall discuss in this review, include C$_3$N, C$_4$N, and C$_5$N.

2.1.2.1 C$_3$N The structure of C$_3$N contains well-defined homogenous rings with no large holes. It is a 2D honeycomb lattice configuration that displays a D$_{6h}$-symmetry that facilitates high structural stability and superior thermal conductivity (Fig. 4g). It is a semiconductor which displays a unique molecular orbital with a low unoccupied molecular orbital gap (~2.7 eV) and an indirect bandgap of 0.39 eV (Fig. 4h) [23].

2.1.2.2 C$_4$N Compared to the atomic structure of another carbon nitride, the C$_4$N structure is filled with sp$^3$ hybridized species and due to the presence of 4 $p_z$ atomic orbitals in the projected electron density of states (PDOS), it exhibits a Dirac cone shape in its band structure like that of graphene (Fig. 4i, j). It is often called the dumbbell (DB) C$_4$N and depending on the positions of the raised C/N atoms in its structure, two structural configurations exist, namely the DB C$_4$N-I and the DB C$_4$N-II. The former refers to a C$_4$N monolayer structure in which the raised C/N atoms are located on the same side, while the latter is a C$_4$N monolayer structure where C/N atoms are on opposite sides [24, 25]. These two configurations of C$_4$N are semiconductors in nature and display a narrow and zero bandgap (Fig. 4k); however, this property can be tuned easily [24, 26].

2.1.2.3 C$_5$N C$_5$N is the most recently reported graphitic-N-based carbon nitrides; like C$_3$N, the atomic structure of C$_5$N is filled with sp$^3$ hybridized carbon atoms; however, it contains two cyclic rings of different sizes (Fig. 4i). A 5-membered carbon ring and an eight-membered carbon ring with two nitrogen atoms are opposite to each other, and these rings are side-by-side to each other (see the atomic structure of C$_3$N in Fig. 4k). The C$_5$N structure has some interesting features such as high chemical, mechanical, and thermodynamic stability. Also, analysis of the band structure shows that the Fermi level is located below the valence-band maximum, which suggests it will be metallic and display superior conductivity than other carbon nitrides (Fig. 4m, n) [27].

2.2 Doped Carbon Nitrides

It has been proven that the metal ion storage capacity of an electrode material such as carbon nitride is significantly dependent on the electronic property. Most carbon nitrides are semiconductors, the electronic density of states (DOS) of the majority of the carbon nitrides possess a lone pair orbital of nitrogen at their Fermi level, and this leads to a large effective mass for hole/electron and results in poor electronic conductivity [19]. Therefore, doping is important for carbon nitride structures. Heteroatoms of elements at the top end of the periodic table such as sulfur, nitrogen, boron, and phosphorus, display narrow bandgap and lower high occupied molecular orbital, which signifies superior electronic conductivity. Therefore, replacing some of the carbon atoms in the crystalline structure of carbon nitrides with metallic or non-metallic atoms can boost their electronic conductivity and electron mobility. Doping also creates surface defects on the carbon nitride structure, and this is beneficial for effective charge transport.

Three heteroatoms are commonly used to dope carbon nitrides. These are carbon, nitrogen, sulfur, and phosphorus. C doping in carbon nitrides can facilitate the formation of delocalized π-bonds, which can result in $n - \pi^*$ electronic transition due to interaction with inherent nitrogen, and this will boost the electronic conductivity and charge transfer. N and S doping have proven to enlarge the interlayer distance of carbon nitrides; this facilitates the adsorption of metal ions and improves the conductivity. Cha et al. reported S-doped mesoporous CN carbon nitride (Fig. 5a shows the optimized structure), and from the electron density profile presented in Fig. 5b, it is obvious that effective charge distribution can be achieved at S-doped sites [31]. Boron (B) and phosphorus (P) doping can decrease the indirect bandgap of semiconductor carbon nitrides and make them metallic. Molaei et al. studied the effect of doping C$_3$N$_2$ with phosphorus by replacing some of the N atoms with P atoms (Fig. 5c), and the DOS
result shown in Fig. 5d confirms a shift of the Fermi level toward the conduction band. This suggests a narrowed band gap and enhanced electronic conductivity compared to pure C3N4 [32]. In summary, heteroatom doping of carbon nitrides is beneficial for boosting their electrical conductivity, improving charge transfer, enlarging their interlayer distance, and boosting metal ion storage.

2.3 CNBCs

Although doping is an effective way of modulating the electronic conductivity of carbon nitrides, they are still limited by poor reversible capacity due to ineffective metal ion storage mechanisms and inadequate surface area. Most carbon nitrides are not able to achieve the desired performance in battery testing because the adsorption energy may be too strong for effective adsorption and desorption of metal ions or the diffusion barrier may be too high for easy metal ion transport. Also, apart from poor electronic conductivity, pure carbon nitrides are characterized by smaller surface area, which provides inadequate coverage for metal ion adsorption. CNBCs can solve these problems because combining pure carbon nitride with highly conductive materials like reduced graphene oxide (rGO) will produce a material with improved electronic conductivity and larger surface area. Moreover, the formation of CNBCs with high theoretical capacity metal oxides such as Fe2O3 will provide an anode material with an improved reversible capacity [33]. CNBCs can also be formed by combining pure carbon nitride with metal chalcogenides, perovskites, etc., for different metal-ion batteries. Hence, we summarize that an effective way to resolve the problems of poor reversible capacity and limited surface area is through CNBCs.
3 DFT-Guided Studies of CNBMs for Energy Storage Devices

After the first discovery/synthesis of tri-s-triazine-based carbon nitride by Berzelius in 1834, the prediction and synthesis of beta-C$_3$N$_4$ in 1993, the discovery of other carbon nitride structures have largely depended on theoretical first-principle simulations. First-principle calculations allow researchers to calculate or predict the structures and properties of material before its synthesis. It can also be used to resolve the discrepancy between experimental measurements and theoretical calculations by considering various possible alkali-metal atom diffusion kinetics, including diffusions in the bulk structure, on the surface, in the defect rich sites, etc. [1, 34]. This prevents random experimental tests or synthesis, thereby facilitating materials discovery. Furthermore, modification strategies for improving the properties of prospective materials can be easily explored through first-principle calculations. DFT is one of the most common computational simulation methods. It is a very powerful tool used to predict different electrochemical parameters of electrode materials. DFT calculations can be used to determine the alkali-metal atom insertion voltage of electrode materials, calculate the migration energy barriers of the alkali-metal atom, and directly visualize the transport pathways and dynamics [35, 36]. In addition, the theoretical capacity of a potential electrode material can be predicted from DFT calculations by investigating the maximum metal atom loading on its structure. The adsorption energy of important molecules on the structure of electrode materials can also be calculated; for example, the adsorption of polysulfides on Li–S cathode materials. Crystal and surface defects are important properties of an electrode material because they impact the alkali-metal atom transport mechanism and pathways, and their effect can also be studied through first-principle calculations. Moreover, information on the intrinsic electronic conductivity and bandgap can be directly inferred from the calculated electronic density of states by observing the Fermi level, conduction, and valence bands. The important role of DFT calculations in CNBMs for energy storage devices includes the DFT-guided synthesis and the DFT-predicted electrochemical properties. In terms of DFT-guided synthesis, DFT studies can determine important synthesis parameters such as the formation energy, exfoliation energy, and cohesive energy of a CNBM. These are criteria that indicate the ease/feasibility of synthesizing a CNBM experimentally. DFT calculations can also provide an atomistic understanding of the interaction between the C/N and the adsorbed metal atom, which will reveal the structural limitations of the CNBMs and guide the synthesis of superior material. DFT studies also play a significant role in predicting fundamental electrochemical properties such as the bandgap, metal atom adsorption energy, open-circuit potential, charge transport kinetics, and theoretical capacity of the CNBMs. These parameters forecast the prospect of CNBMs for energy storage devices and reveal potential limitations that must be considered and improved upon. The electrochemical properties that can be predicted from DFT are summarized in Fig. 6.

3.1 Theoretical Calculations on CNBMs for Energy Storage Devices

Density functional theory calculations are one of the most useful tools for discovery and electrochemical investigation of different carbon nitrides for rechargeable batteries [41]. In this section, we discuss different carbon nitrides that have been studied through DFT.

3.1.1 Pure Carbon Nitrides

Through DFT calculations, it was predicted that C$_3$N$_4$ carbon nitride could deliver a theoretical capacity of up to 524 mAh g$^{-1}$ (Li$_2$C$_3$N$_4$) [42], and this capacity has not been achieved in battery testing. Mao et al. [43], Veith et al. [44], and Hankel et al. [45] showed that this poor performance was due to the high content of graphitic nitrogen in its structure which resulted in ineffective intercalation. We reported the first C$_3$N$_4$ with reduced graphitic-N and increased pyridinic-N for LIBs, and our DFT calculations showed that unlike bulk/sheet C$_3$N$_4$ which adsorbs Li atom in the triangular pore at high energy of ~ 4 eV, a 1D-C$_3$N$_4$ fiber possesses lower Li binding energy (2.61 eV) at its edges. Hence, it can effectively adsorb/desorb Li atoms (Fig. 7a, b) [46]. Hui Pan reported a first-principle study on C$_3$N$_4$ carbon nitride nanotubes for LIBs and predicted that it could store Li atoms internally and externally due to its porous structure, which might make it better than other dimensions of carbon nitride [47].
Other carbon nitride structures have also been studied for LIBs through DFT. For example, Hankel et al. showed that although g-CN can deliver a high capacity of 454 mAh g\(^{-1}\). Its high Li binding energy of > 3 eV leads to ineffective intercalation/deintercalation and limits its prospects for LIBs [48]. The study of Hussain et al. on C\(_2\)N-h2D showed that despite the pyridinic nitrogen, which facilitates Li storage, the high initial Li adsorption energy would result in poor battery performance [16]. By using a “pair of particle (metals) model,” Wu et al. [49] carried out a DFT study on C\(_2\)N for LIBs and SIBs to show that C\(_2\)N monolayer can achieve a capacity of 2939 and 2469 mAh g\(^{-1}\) for LIBs and SIBs with low diffusion barrier and an OCV of 0.45 V [16]. Liu et al. carried out a DFT study on different C\(_3\)N compositions (C\(_3\)N, C\(_{2.67}\)N, and C\(_{3.33}\)N) and showed that the drop in capacity experience by Xu et al. [50] was due to ineffective intercalation of Li. They concluded that C\(_{3.33}\)N is the preferred C\(_3\)N composition, and it delivered a reversible capacity of 840.35 mAh g\(^{-1}\) (Fig. 7c), operated at a low open-circuit potential of 0.12 V, and displayed superior electronic conductivity [40]. Guo et al. proposed other C\(_3\)N allotropes (C\(_3\)N-S1, C\(_3\)N-S2, and C\(_3\)N-S3) with unique electronic properties, and the C\(_3\)N-S2 structure displayed the most feasible Li diffusion pathway with lower diffusion barrier (0.14 eV) for LIBs (Fig. 7d, e) [51]. Yang et al. also reported the first-principle study of two structural configurations of DB C\(_4\)N as LIBs anode for LIBs, and they predicted that DB C\(_4\)N-I and DB C\(_4\)N-II could deliver high theoretical capacities of 1942 mAh g\(^{-1}\) (DB C\(_4\)N-I) and 2158 mAh g\(^{-1}\) (DB C\(_4\)N-II) [52].

DFT studies of pure carbon nitrides for large-sized metal ion batteries have also been reported. For instance, Weng et al. showed that pure C\(_3\)N\(_4\) nanosheet displays high adsorption energy for Na-atom, but it shows an exceedingly high diffusion barrier, which makes it fail as a SIBs anode [53]. Moreover, through DFT we showed that thanks to the high pyridinic-N of
Fig. 7 Reversible Li active sites in a 2D-C₃N₄ sheet, and b 1D-C₃N₄ fiber. Green means reversible Li atoms and red means non-reversible Li atoms. c Top and side view of stable intercalation structures of n Li ions into C₃N. Reproduced with permission from Ref. [40]. d Diffusion paths and e corresponding energy barriers of Li migration in C₃N-S₂. Reproduced with permission from Ref. [51]. f Binding energy of Li₂S₄/Li₂S₆/Li₂S₈ interacting with G, BN, C₃N, C₃N₄, and DOL/DME solvent, respectively. Reproduced with permission from Ref. [17]. g Isosurfaces of charge density difference of Li₂S, Li₂S₂, Li₂S₄, Li₂S₆, Li₂S₈, and S₈ adsorbed on the surface of C₃N with the isovalue of 0.003 Å⁻³. Blue wire-frames denote loss of electrons and yellow wireframes denote gain of electrons. Reproduced with permission from Ref. [27]. Copyright permissions from American Chemical Society, Elsevier and Wiley–VCH. (Color figure online)
1D-C3N4 fiber it exhibits a high affinity for potassium ions, but it suffers from a rather high K-diffusion barrier which limits effective ion transport [54]. Bhuriyal et al. also reported the first-principle study of C3N monolayer as a promising anode material for SIBs and PIBs, and they showed that multilayer adsorption of the metal atoms generated a high capacity of 1072 mAh g−1 and low diffusion barrier of 0.03 and 0.07 eV for Na and K-ion [55]. Xu et al. showed that metal ion (Li, Na, and K) adsorption on C4N is an effective way to open the zero bandgap and modulate the electronic property because the adsorbed metal ion transfers charges to the surface of DB C4N [30].

Thanks to the nitrogen-rich structure, carbon nitrides have also been studied for Li–S and other battery systems. Li et al. reported that due to the accumulation of charges at the N–N bond of the C4N4 structure, it demonstrates superior lithium polysulfide species (LIPSs) anchorage than commercial electrolyte solvent molecules (DOL and DME) [56]. Zheng and coworkers compared the LIPSs anchoring property of graphene, boron nitride (BN), C2N, and C3N4 with commercial solvents and showed that C2N and C3N4 were the most effective due to their stronger binding energy (Fig. 7f) surface interaction with LiPSs via the Li-N/C-S bonds formed during their interaction [17]. Liang et al. also showed that interaction between LIPSs and polymeric C3N4 (p-C3N4) exhibits strong ionic bonding, electrostatic, and vdW interactions which are beneficial for altering the bonding and spatial configuration of LIPSs which then modifies their redox kinetics [57]. Meng et al. also reported that the abundant nitrogen species on the surface of C3N4 nanosheet is able to facilitate LiPSs anchoring by a surface chemical adsorption mechanism due to the formation of a Li-N bond [58]. Wang et al. reported a first-principle study on the application of C3N as a LIPSs host. The isosurfaces of charge density difference presented in Fig. 7g showed that it exhibits an effective physical/chemical adsorption property, which enables LiPSs anchoring and charge transfer to its surface [27]. Carbon nitrides have also been explored for metal-air batteries because of their unique properties. For example, Shinde et al. reported the prospect of C2N for zinc-air (Zn-Air) batteries and showed that the open holey structure of C2N enabled reversible oxygen reactions and improved the electronic conductivity [59]. Je et al. also carried out first-principle calculations on heptazine and triazine sourced C3N4 for non-aqueous Li-O2 battery, and the results showed that triazine was better because it delivered a higher overpotential. Interaction of C3N4 with LiO2 resulted in the formation of a Li-N bonding, which is dependent on the ratio of N in the material and greatly influences their overpotential [60].

### 3.1.2 Doped Carbon Nitrides

Theoretical study on doped carbon nitrides of different atomic compositions has been reported. For example, Nong et al. considered the effect of non-metal doping (boron, oxygen, and sulfur) and the impact of strain on the Li storage capacity and Li absorptivity of C3N. They reported that oxygen-doped C3N (Ox–C3N) was the preferred dopant, it operated at 0.02 V and delivered a higher theoretical capacity of 534.42 mAh g−1 although it exhibited a high Li diffusion barrier of 0.78 eV, and this could be lowered by applying strain to the C3N structure [61]. Tian et al. also studied the effect of boron doping on the performance of C3N for alkali metal ion battery (Li, Na, and K) and showed that B4-doped C3N displays superior capacity, cycle, and thermal stability than pristine C3N [29]. Cha et al. showed that CN possesses a high Na adsorption energy, which can foster attraction of Na atoms into its structure for SIBs batteries [31]. Although Weng et al. [62] proved that C3N4 displays a very high Na-adsorption energy which limits its application for SIBs, the report of Molaei et al. on P-doped C3N4 showed that phosphorus doping is an effective way to decrease the high Na adsorption energy and diffusion barrier in C3N4 (Fig. 8a-d). The more P-atoms introduced into the structure of C3N4, the lower these energies [32]. Moreover, the work of Cha et al. [31] on Si-doped CN suggests that S-doping of carbon nitrides can successfully enlarge their interlayer distance for such large-sized metal-ion battery applications.

Doped carbon nitrides have also been studied for Li–S battery, Lin et al. studied the effect of transition metal doping (Mn, Fe, Co, Ni, and Cu) in C3N for improved immobilization of polysulfides in Li–S batteries, their results showed that a Lewis acid–base interaction which boosted LiPSs anchoring was established, and a significant charge was transferred from the transition metal dopant to the C2N monolayer with Co@C2N transferring the most in line with its superior adsorption energy (Fig. 8e, f) [28]. Zhao and coworkers reported confined single-atom Pt in holey g-C3N4, and DFT calculations showed that incorporation of single-atom Pt in holey g-C3N4 leads to improved electrical
conductivity and a more stable structure with efficient electron and ionic transfer [63].

3.1.3 CNBCs

CNBCs with superior electronic conductivity and higher capacity have also been studied through DFT. For example, Wang et al. reported a DFT study of C\textsubscript{2}N/graphene heterostructure for LIBs and proved that the electrical conductivity and structural stability can be significantly improved. The heterostructure recorded a high theoretical capacity of 1079 mAh g\textsuperscript{-1}, a low Li diffusion barrier of 0.28 eV at the interlayer (Fig. 9a), and operated at a low open-circuit voltage of 0.13 V [37]. Ding et al. reported the first-principle study of C\textsubscript{2}N/graphene bilayer for LIBs, and by using molecular dynamic (MD) simulations, they predicted that Li storage follows a two-step process, i.e., migration through the z-direction via the large hole in the center of the C\textsubscript{2}N...
structure and on to the surface of the C$_3$N membrane. The structures of different compositions of the bilayer heterostructures are presented in Fig. 9b, c. [38]. Guo et al. [64] and Lin et al. [65] also reported a DFT study on C$_3$N/phosphorene heterostructure for LIBs, while Bao et al. explored its potential for Li/Na battery. The three reports concluded that the design of such C$_3$N/phosphorene heterostructure would not only address the issues of phosphorene but also alleviate the problems of C$_3$N for Li/Na storage. Storage of Li-ions occurred at the outer surface and interlayer of the heterostructure (Fig. 9d, e) [66].

DFT reports on CNBCs for Li–S batteries have also been widely reported. For instance, Liao and co-workers showed that the anchoring ability of C$_3$N$_4$ could be exploited in CNBCs [67]. Fan et al. also reported a carbon black blended g-C$_3$N$_4$ (g-C-coated) for Li–S battery and showed that two bonds (C–S and N–Li) are formed as a result of its interaction with LiPSs. These bonds inhibit the migration of LiPSs because it effectively binds to the LiPSs [68]. The work of Li et al. on porous C$_4$N$_4$ monolayer and C$_3$N$_4$/graphene composite for Li–S battery reported that LiPSs adsorption occurred by a chemisorption process and addition of graphene enhanced this process (Fig. 9f) [56]. Chen et al. studied the effect of transition metal (Fe, Ni, Cu, and Co)-doped g-C$_3$N$_4$/C composite for Li–S battery and showed that this would increase the electron mobility, while g-C$_3$N$_4$ will prevent polysulfide dissolution by anchoring LiPSs species [69]. In summary, DFT has been very key in the synthesis and electrochemical study of carbon nitrides for metal-ion
batteries. Table 1 compares the DFT predicted parameters of carbon nitrides with commercial graphite anode for energy storage devices. From this table, it is obvious that most of the carbon nitrides can function as anode materials with superior theoretical capacity compared to graphite. These electrochemical properties have inspired researchers into the synthesis and fabrication of pure and doped carbon nitrides as well as CNBCs for energy storage devices.

### Table 1: Comparison of the DFT predicted parameters of pure carbon nitrides with commercial graphite for energy storage devices

| Materials     | Conductivity | Capacity (mAh g⁻¹) | Adsorption energy (eV) | Diffusion barrier (eV) | Operating voltage (V) |
|---------------|--------------|---------------------|------------------------|------------------------|------------------------|
| Graphite      | Metallic     | 372 (LIBs)          | 1.1–1.29 (LIBs) [72]   | 0.22 (LIBs) [75]       | 0.0–0.5 (LIBs) [78]     |
| CN            | Semi-conductor | 454 (LIBs) [48]    | – 5.03 (LIBs) [48]     | ~ 3 (LIBs) [45]        | ~ 0.6 (LIBs) [81]       |
| C₂N           | Semi-conductor | 2939 (LIBs) [49]   | – 3.437 (LIBs) [49]    | 0.409 (LIBs) [49]      | 0.452 (LIBs) [49]       |
| C₃N           | Semi-conductor | 2469 (SIBs) [49]   | – 2.868 (SIBs) [49]    | 0.116 (SIBs) [49]      | 0.458 (SIBs) [49]       |
| C₃N₄          | Semi-conductor | 837.06 (LIBs) [40] | – 0.01 (LIBs) [40]     | 0.8 (LIBs) [40]        | 0.15 (LIBs) [40]        |
| C₅N           | Semi-conductor | 524 (LIBs) [44]    | – 4.56 (LIBs) [45]     | ~ 1.8 (LIBs) [47]      | 0.8 (LIBs) [44]         |
| C₆N           | Semi-conductor | 1072 (SIBs) [55]   | – 1.806 (SIBs) [55]    | 0.03 (SIBs) [55]       | 0.13 (SIBs) [55]        |
| C₅C₂N         | Semi-conductor | 1072 (PIBs) [55]   | – 2.230 (PIBs) [55]    | 0.07 (PIBs) [55]       | 0.26 (PIBs) [55]        |
| Melamine      | Melange      | N/A                 | N/A                    | N/A                    | N/A                    |
| Cyanamide     | Melange      | N/A                 | N/A                    | N/A                    | N/A                    |
| Dicyandamide  | Melange      | N/A                 | N/A                    | N/A                    | N/A                    |
| Urea          | Melange      | N/A                 | N/A                    | N/A                    | N/A                    |
| Thiourea      | Melange      | N/A                 | N/A                    | N/A                    | N/A                    |

4 **Synthesis Strategies of Pure and Doped Carbon Nitrides**

The conclusions obtained from DFT studies showed that the poor performance of most CNBMs can be traced to high metal atom adsorption energy due to excess graphitic-N, which leads to irreversible intercalation/deintercalation, poor conductivity, and low charge transfer mobility, as well as inferior structural stability after metal atom adsorption. DFT calculations showed that these problems could be overcome by regulating the ratio of C/N in the structure and prioritizing pyridinic-N, heteroatom doping,

---

**Fig. 10**  
(a) Schematic illustration of one of the top-down synthesis approach (thermal polymerization) for g-C₃N₄ using different precursors. Black balls—carbon (C), blue balls—nitrogen (N), white balls—hydrogen (H), red balls—oxygen (O), and yellow balls—sulfur (S), respectively. Reproduced with permission from Ref. [84].  
(b) Schematic of the synthesis of g-C₃N₄ nanosheets from bulk g-C₃N₄. In the atomic model, carbon atoms—gray balls, nitrogen atoms—blue balls and hydrogen atoms—red.  
(c) TEM image of g-C₃N₄ nanosheets. d Tapping-mode AFM image of a single g-C₃N₄ nanosheet deposited on the silicon wafer substrate. The inset is the height curve determined along the line between P1 and P4. Reproduced with permission from Ref. [88]. Copyright permissions from American Chemical Society and Wiley–VCH. (Color figure online)
and CNBCs design with other superior electrode materials. Inspired by these DFT conclusions, several researchers have modified the general experimental synthesis strategies to obtain CNMBs that exhibit the desired structural/electronic properties. Such modified experimental synthesis strategies that are motivated by the findings in DFT studies can be considered DFT-guided synthetic protocols. In this section, we summarize some significant DFT-guided synthesis strategies for CNBMns, including pure/doped carbon nitrides as well as CNBCs.

4.1 Top-Down Strategy

Carbon nitrides generally occur in the bulk state, and this form of carbon nitride is obtained via thermal decomposition of N-rich amine compounds such as melamine [82] (Fig. 10a). However, bulk carbon nitride suffers from the poor conductivity, limited surface area, and sluggish reaction kinetics [83, 84]. Bulk carbon nitride also contains excess graphitic-N which is proven to limit the performance of carbon nitrides in rechargeable batteries according to DFT conclusions. One way to regulate the C/N ratio and decrease the graphitic-N is by converting bulk carbon nitride to nanosheets and other morphologies. This can be achieved through the top-down strategy which is grouped into post-thermal oxidative etching and ultrasonic liquid exfoliation.

4.1.1 Thermal Oxidation

As the name implies, this method involves the heat treatment of bulk carbon nitride at high temperature and it is an effective way to break the van der Waals forces which hold the layer of carbon nitride together. This process produces a sheet-like carbon nitride with large surface area, surface defects and adjusted layer thickness [22, 85]. As depicted in Fig. 10b, bulk g-C$_3$N$_4$ can be converted to g-C$_3$N$_4$ nanosheets via thermal oxidation and Fig. 10c, d shows that the layer thickness can also be regulated. Guo et al. reported the synthesis of graphite/sheet-like carbon nitride (CN) as well as nanotube, nanoribbon, and microsphere morphologies via thermal treatment [86, 87]. 2D-C$_3$N carbon nitride with a needle-like morphology was also synthesized by a direct solid-state reaction [23].

4.1.2 Ultrasonic Liquid Exfoliation

Another effective strategy to convert bulk multilayered carbon nitrides to few/single layer carbon nitrides is through the intercalation of solvent molecules through the bulk structure, thereby resulting in delamination (Fig. 11a). This liquid exfoliation process is effective for synthesizing nanosheets or flakes of carbon nitrides (Fig. 11b, c). Mahmood et al. reported the synthesis of two-dimensional C$_2$N carbon nitride by a top-down (facile wet-chemical
reaction) approach [89]. This process can also be achieved through lithiation exfoliation which uses Li ions as exfoliant to penetrate the layers of bulk carbon nitride and separate them to individual nanosheets (Fig. 11d).

4.2 Bottom-Up Strategy

The bottom-up strategy or template-aided synthesis is divided into soft and hard templating method.

4.2.1 Hard Templating

Templates are often used as morphology directing agents during the synthesis of unique carbon nitride morphologies. Hard templates such as silica [93–95], anodic alumina oxide [96], and carbon [97] are often reported for synthesizing CNBMs. For instance, DFT studies showed that by increasing the N content in C$_3$N$_4$ to C$_3$N$_5$ the electrochemical properties can be improved. Recently, Kim et al. reported the synthesis of a graphene-like mesoporous carbon nitride C$_3$N$_5$ with superior electronic properties by using KIT-6 as a hard template [98]. Moreover, their DFT/electrochemical study showed that such composite can deliver excellent performance for energy storage devices [34]. The C$_3$N$_5$ sample showed outstanding interesting electrochemical properties, and it was combined with graphene oxide to form a composite (MCN-11). Figure 12a shows a scheme of the synthesis procedure of the C$_3$N$_5$ and the hybrid.

4.2.2 Soft Templating

Templating materials which demonstrate self-assembling properties have also been employed for synthesizing carbon nitrides by using ionic liquids [99, 100], surfactants [101–103], and amphiphilic block polymers [102, 104]. Yong et al. reported the synthesis of a nanoporous carbon nitride by using ionic liquid as soft templates (Fig. 12b-e), and such carbon nitride structure demonstrated superior conductivity to bulk material [101]. In summary, by using the conclusions of DFT studies as a guide, the general experimental synthesis approach of carbon nitrides can be optimized to achieve carbon nitrides with regulated C/N ratio, increased pyridinic-N, superior structural stability, and conductivity for improved performance.

4.3 Fabrication Strategies of Doped Carbon Nitrides

The DFT study of Molaei et al. [32] proved that heteroatom doping of carbon nitride will boost the electronic conductivity and improve metal atom storage, and the DFT conclusion of this work motivated the Vinu group to report the synthesis of rod-like sulfur-doped mesoporous CN (S-CN) by a templating method (Fig. 13a) [31]. Phosphorus-doped mesoporous carbon nitride (P-MCN) was also synthesized via a simple template approach [105]. A schematic of the template synthesis method for heteroatom-doped mesoporous carbon nitrides is presented in Fig. 13b. To summarize, doped carbon nitrides can be synthesized through a combination of some top-down and bottom-up approaches.
which often involves the use of a salt of the heteroatom or a surfactant.

5 Fabrication Strategies for CNBCs

DFT studies have proved that CNBCs design is another effective way to resolve the challenges of carbon nitrides because such CNBCs will demonstrate superior conductivity, better structural stability, and enhanced charge transfer. Therefore, in this section, we highlight some recent CNBCs that have been synthesized through commonly reported experimental synthesis strategies but inspired by the conclusions of DFT studies.

5.1 Hydrothermal Method

Hydrothermal method involves the controlled synthesis of a composite in a tightly sealed vessel under high temperature and pressure [106, 107]. Although 2D-C₃N₄ is widely reported for designing CNBCs, our DFT studies showed that 1D-C₃N₄ exhibits superior structural stability and metal storage capability than 2D-C₃N₄ [40]. Moreover, our comparative DFT study between 1D/2D C₃N₄/graphene and 1D/2D C₃N₄/rGO showed that 1D/2D will perform better [38]. These DFT conclusions motivated us to combine 1D-C₃N₄ with 2D-rGO for energy storage devices. Therefore, we reported the synthesis of a 1D/2D C₃N₄/rGO composite via

Fig. 13  a Schematic of the synthesis procedure of S-MCN [31]. b Schematic of the template synthesis method for heteroatom-doped mesoporous carbon nitrides. Reproduced with permission from Ref. [105]. Copyright permission from American Chemical Society. (Color figure online)
a freeze-drying-assisted hydrothermal approach by deploying the π–π interaction between C₃N₄ and graphene [54]. Figure 14a shows the synthesis scheme of the composite. Guided by the result of our DFT calculations, we also reported the design of a composite of Co₃O₄@N–C derived from 1D-C₃N₄ via an ionic liquid-assisted solvothermal method [108]. Scheme illustration of the synthesis strategy is depicted in Fig. 14b.

5.2 Self-Assembly Method

Due to their amphoteric nature, carbon nitrides possess tunable surface functional groups and surface charges which enable surface attachment with other functional materials such as graphene and other functional 2D materials [130]. Such structural interaction has been identified through the molecular dynamic simulation and DFT studies of Ding et al. and Wang et al. The heterostructure will display superior electronic conductivity and structural stability than the pure carbon nitride. This concept is employed in self-assembly synthesis of C₃N₄ and other CNBMs. Figure 15 shows a detailed schematic of the self-assembly approach for a surface modified C₃N₄ (pCN) with graphene oxide and reduced graphene oxide to achieve a GO/pCN and rGO/pCN composite, respectively [109].

5.3 Other Composite Fabrication Strategies

CNBCs can also be designed by some other unique methods, and Fu et al. designed a 2D-2D g-C₃N₄-rGO hybrid via an in situ chemical method by initiating a nucleophilic reaction between epoxy groups on the surface of GO and amine/amide groups on dicyanamide-C₂H₄N₄. The synthesis method is depicted in Fig. 16a, and through this approach, the problem of poor electrical conductivity, aggregation, and restacking was resolved [110]. Similarly, Li et al. reported the design of a Zn₂GeO₄/g-C₃N₄ composite by growing Zn₂GeO₄ NPs in-between the layers of g-C₃N₄ via a solution approach to inhibit agglomeration of Zn₂GeO₄ NPs and restacking of g-C₃N₄ (Fig. 16b). Zn₂GeO₄ also functioned as a spacer to enlarge the interlayer distance of g-C₃N₄ sheet enabling metal ion adsorption and improved conductivity [111]. Wang et al. reported the synthesis of a GO/g-C₃N₄ microsphere by an ethanol-assisted spray drying approach [112]. Zhang et al. also reported the design of a sponge-like free-standing 3D S/graphene@g-C₃N₄ hybrid using a microemulsion-assisted assembly method (Fig. 16c) [113].
Fig. 15 Schematic diagram for the synthesis process of rGO/pCN samples via a combined ultrasonic dispersion and electrostatic self-assembly strategy followed by a NaBH₄-reduction process. Reproduced with permission from Ref. [109]. Copyright permission from Elsevier. (Color figure online)
In summary, the advancement in experimental synthesis strategies of pure/doped carbon nitrides and CNBCs has been propelled by the conclusions of DFT studies on CNBMs. Precisely, DFT studies proposed that adjusting the C/N ratio, increasing the concentration of pyridinic-N, heteroatom doping and CNBCs design are effective ways to inhibit the problem of irreversible intercalation/deintercalation, poor conductivity, and instability experienced by pure layered CNBMs in rechargeable batteries. These conclusions from DFT studies have guided the experimental synthesis of CNBMs to fabricate functional CNBMs which exhibit the desired requirements and deliver superior battery performance.

6 Electrochemical Studies of CNBMs for Energy Storage Devices

The structural/electronic properties and surface functionalities of CNBMs qualify them as promising electrode materials for energy storage devices. In this section, we give an overview of experimental works on carbon nitrides for energy storage devices including LIBs, SIBs and PIBs, Li–S, LABs, LMBs, ZABs, and SSBs.

6.1 Lithium-Ion Batteries (LIBs)

LIBs offer benefits such as lightweight, superior energy density, and long cycle life [3, 114, 115]; CNBMs (pure/doped carbon nitrides and CNBCs) have been reported as electrodes.

6.1.1 Pure Carbon Nitrides for LIBs

Due to the structural and electronic properties of carbon nitrides, different atomic composition of carbon nitrides has been studied for LIBs. Yin and co-workers reported C$_3$N$_3$ for LIBs but only a low reversible capacity of 197.8 mAh g$^{-1}$ was obtained at 100 mA g$^{-1}$ after 300 cycles and a ICE of 34.3% [81]. Xu et al. tested C$_2$N (C$_2$N-450) and C$_3$N for LIBs and at 1C, C$_3$N only retained 285.1 mAh g$^{-1}$ after 500 cycles, while C$_2$N-450 maintained a reversible capacity of 516.1 mAh g$^{-1}$ although it displayed large activation (Fig. 17a) [50]. Other carbon nitride compositions such as C$_4$N and C$_5$N have not been experimentally tested for any metal ion batteries.

Fig. 16  a Illustration of the formation process of g-C$_3$N$_4$–rGO. Reproduced with permission from Ref. [110]. b Schematic of the synthesis process of the Zn$_2$GeO$_4$/g-C$_3$N$_4$ hybrids. Reproduced with permission from Ref. [111]. c Schematic illustration of the procedure for preparing S/GCN hybrid sponge. Reproduced with permission from Ref. [113]. Copyright permissions from Royal Society of Chemistry and Wiley–VCH. (Color figure online)
We reported the synthesis of a porous 1D-C$_3$N$_4$ fiber with large surface area and multiple active sites (see SEM image—Fig. 17b) for LIBs; a reversible capacity of 181.7 mAh g$^{-1}$ was achieved at 0.5 C and 138.6 mAh g$^{-1}$ at 10 C (Fig. 17c) [46]. Chen et al. also reported the application of C$_3$N$_4$ (ND-g-C$_3$N$_4$) for LIBs and obtained a capacity of 2753 mAh g$^{-1}$ after 300 cycles) although significant activation occurred (Fig. 17d). To provide a brief summary, pure carbon nitrides of different crystalline structures have been reported for LIBs, and commendable performances have been recorded (Table 2). However, the issues of ineffective intercalation/deintercalation due to extremely high Li adsorption energy, high Li diffusion barrier, structural deformation, and loss of crystallinity have severely limited the exploitation of their full potential. Therefore, DCNs and CNBCs are necessary to achieve improved performance.

Fig. 17 a Long cycle life of C$_2$N-450, C$_3$N. Reproduced with permission from Ref. [50]. b SEM image of the 1D-g-C$_3$N$_4$ fiber. c Cycling performance of 1D-g-C$_3$N$_4$ fiber structure at a high current density of 10 C. Reproduced with permission from Ref. [46]. d Rate performance for ND-g-C$_3$N$_4$ electrode at current density of 0.1, 0.2, 0.5, 1, 2, 5, 10, and 20 A g$^{-1}$ and galvanostatic discharge property for g-C$_3$N$_4$ electrode. Reproduced with permission from Ref. [8]. Copyright permissions from Wiley–VCH, and American Chemical Society. (Color figure online)
6.1.2 CNBCs for LIBs

CNBCs can deliver superior structural/electronic properties, enhanced structural stability and extremely large surface area which will boost the overall LIBs performance. In this section, we highlight some significant CNBCs including binary CNBCs and ternary CNBCs.

6.1.2.1 Binary CNBCs for LIBs

Binary CNBCs involving carbonaceous materials such as graphene oxide are commonly reported because of the inherent π–π stacking, extremely large surface area, surface functional groups, and electronic conductivity. For instance, Fu et al. [110] reported the application of a 2D-2D stacked g-C\textsubscript{3}N\textsubscript{4}-rGO hybrid for LIBs. The 2D-2D stacked g-C\textsubscript{3}N\textsubscript{4}-rGO hybrid achieved a reversible capacity of 1525 mAh g\textsuperscript{−1} at 100 mA g\textsuperscript{−1} and up to 943 mAh g\textsuperscript{−1} at 1000 mA g\textsuperscript{−1} [111]. Mohamed et al. designed a CuO/O-doped g-C\textsubscript{3}N\textsubscript{4} composite which delivered a reversible capacity of 250 mAh g\textsuperscript{−1} at 30 mA g\textsuperscript{−1}, 50 mAh g\textsuperscript{−1} at 50 mA g\textsuperscript{−1} and 181.7 mA h g\textsuperscript{−1} at 200 mA g\textsuperscript{−1} [46].

6.1.2.2 Ternary CNBCs for LIBs

Ternary composites which can provide additional benefits than binary composites have also been reported. Wang et al. reported the application of a Si@rGO/g-C\textsubscript{3}N\textsubscript{4} composite for LIBs. By taking advantage of the interfacial chemical bonding on functionalized rGO/g-C\textsubscript{3}N\textsubscript{4}, Si nanoparticles (NPS) were effectively anchored on 2D-composite of rGO/g-C\textsubscript{3}N\textsubscript{4} (Fig. 19a), and such composite design resulted in a reversible capacity of 1354.8 and 799.6 mAh g\textsuperscript{−1} when cycled at 0.1 and 0.5 C, respectively [118].

### Table 2: Pure and doped carbon nitrides for LIBs and SIBs

| Battery type | Materials | Synthesis | Morphology | Initial capacity (mAh g\textsuperscript{−1}) | ICE\textsuperscript{a} | Reversible capacity (mAh g\textsuperscript{−1}) | References |
|--------------|-----------|-----------|------------|---------------------------------|----------------|---------------------------------|------------|
| LIBs         | ND-g-C\textsubscript{3}N\textsubscript{4} | Magnesiothermic denitriding method | Porous nanosheet | 2627 @ 100 mA/g | 45.7% | 2753 @ 300 cycles | [8]         |
|              | CN-480–600 | Solid-state Wurtz reaction | Macroporous graphite-like | 575.7 @ 100 mA/g | 19.9% | 197.8 @ 300 cycles | [144]      |
|              | C\textsubscript{2}N-450 | Bottom-up wet-chemical reaction. | Stacked sheet | 1629.6 @ 0.1 C | 57.2% | 516.1 @ 500 cycles | [146]      |
|              | C\textsubscript{2}N | Bottom-up wet-chemical reaction. | Rod-like | 787.3 @ 0.1 C | 48.7% | 1C, 285.1 @ 500 cycles | [126]      |
|              | Li-C\textsubscript{2}N\textsubscript{4} | Electrochemical and solid-state reactions | Sheet-like | 188 | – | 38 @ 6 cycles | [121] |
|              | C\textsubscript{2}N\textsubscript{4} (C\textsubscript{3}N\textsubscript{4}) | Thermal oxidation | – | 250 @ 30 mA/g | – | 50 @ 50 cycles | [122] |
|              | 1D-C\textsubscript{3}N\textsubscript{4} fiber | Poladdition/polycondensation reaction | Porous layered fiber | 419.7 @ 0.5 C | 84.1% | 181.7 @ 200 cycles | [46]      |
|              | NGC | Carbonization | Graphene-like | 2749 @ 50 mA/g | 50.3% | 1143 @ 200 cycles | [145] |
|              | P-MCN-1 | Template synthesis | Spherical particles | 2850 @ 1 A/g | 44.2% | 963 @ 1000 cycles | [105]     |
| SIBs         | C\textsubscript{2}N\textsubscript{4} (C\textsubscript{3}N\textsubscript{4}) | Thermal oxidation | – | 250 @ 30 mA/g | – | 10 @ 50 cycles | [122] |
| SIBs         | S-MCN | Hard template approach | Rod-like morphology | -850 @ 100 mA/g | 47% | 304.2 @ 100 cycles | [31]      |

\textsuperscript{a}ICE Initial coulombic efficiency
cycles along with a stable rate performance exceeding that of Fe$_2$O$_3$/G [120] (Table 3).

Ternary CNBCs involving metal chalcogenides such as MoS$_2$ and SnS$_2$ have also been studied. Hou et al. reported a C$_3$N$_4$/NRGO/MoS$_2$ composite which exhibited a reversible capacity of 855 mAh g$^{-1}$ after 100 cycles when tested at 100 mA g$^{-1}$ (Fig. 19c) [121]. Shah et al. reported the design of a SnS$_2$, rGO and g-C$_3$N$_4$ composite with intimate 2D-2D contact for LIBs. A stepwise detail of the synthesis method and the reaction processes involved is depicted in Fig. 19d. A reversible capacity of 1248.4 mAh g$^{-1}$ was retained after 276 cycles at 100 mA g$^{-1}$ [122]. Shi et al. also prepared a rGO/g-C$_3$N$_4$@SnS$_2$ composite with effective surface contact of all constituents which can be seen from the TEM image (Fig. 19e). After 1000 cycles at 800 mA g$^{-1}$, a reversible capacity of 864.9 mAh g$^{-1}$ was retained [123].

6.2 Sodium and Potassium-Ion Batteries (SIBs and PIBs)

Due to their large-sized ions, one of the major focus for sodium and potassium ion battery electrode materials is the enlargement of their interlayer distance. The low interlayer/d-spacing of graphite (0.334 nm) and its low theoretical capacity (279 mAh g$^{-1}$) has limited its application in either of these two battery systems [125–127]. g-C$_3$N$_4$ can be a source of N-doped carbon with large interlayer distance; this approach was reported by Qiao et al. who successfully fabricated a series of N-doped graphene 2D-sheet with different interlayer distances (Fig. 20c). The optimized N-doped graphene sheet (N-FLG-800) delivered a superior rate capability of 56.6 mAh g$^{-1}$ at a current density of 40 A g$^{-1}$ (Fig. 20e) and outstanding long-cycle stability of 211.3 mAh g$^{-1}$ after 2000 cycles at 0.5 A g$^{-1}$ [128]. Weng et al. reported the synthesis of a C/g-C$_3$N$_4$ composite via a simple one-pot synthesis approach, and the composite delivered a capacity of 254 mAh g$^{-1}$ at 0.1 A g$^{-1}$ and 160 mAh g$^{-1}$ at 0.4 A g$^{-1}$ (see Fig. 20g) [53]. Recently, Chen et al. showed that coating the surface of copper metal current collector with a thin film of 2D-C$_3$N$_4$ can improve Na storage by inhibiting unwanted surface interaction with the liquid electrolyte. The Na$^+$ storage mechanism is illustrated by the scheme in Fig. 20h. At an areal current density of 0.013 mA cm$^{-2}$, a high areal capacity of 0.036 mAh cm$^{-2}$ was achieved [129].
In the case of potassium-ion batteries (PIBs), we showed that the high K⁺ affinity of 1D-C₃N₄ fiber facilitates high initial PIBs capacity, but it suffers from poor cycle life and DFT study showed that this was due to the high potassium diffusion barrier and poor conductivity. Guided by this theoretical discovery, we combined 1D-C₃N₄ fiber with rGO and fabricated a 1D/2D C₃N₄/rGO composite (Fig. 21a—SEM) which exhibited a larger surface area, superior K⁺ diffusivity, and improved conductivity. The composite delivered a superior reversible capacity 557.4 mAh g⁻¹ after 50 cycles with impressive cycle stability (Fig. 21b). The K-ion storage mechanism is depicted in Fig. 21c and shows the bi-directional transport of ions through the interlayer of the composite [54]. In our recent work, we resolved the inadequate interlayer spacing of Co₃O₄ by coating it with N-doped carbon sourced from 1D-C₃N₄ fiber. The Co₃O₄@N–C possessed a core–shell morphology (Fig. 21d) in which N–C at the surface of Co₃O₄ ensured effective transfer of attracted K⁺ to the Co₃O₄ core. The composite delivered a superior reversible capacity of 448.7 mAh g⁻¹ unlike pure Co₃O₄ spheres which only recorded ~ 10 mAh g⁻¹ after 40 cycles (Fig. 21e). The potassium storage mechanism of the composite is depicted in Fig. 21f.
| Battery type | Materials | Synthesis | Morphology | Initial capacity (mAh g\(^{-1}\)) | ICE\(^{\circ}\) | Reversible capacity (mAh g\(^{-1}\)) | References |
|--------------|-----------|-----------|------------|----------------------------------|------------|-----------------------------------|------------|
| **LIBs**     | huCP/g-C\(_{3}\)N\(_{4}\) | Hydrothermal treatment | Carbon fibers with large pores and hollow structure | 1199 @ 1 A/g | ~ 93% | 1030 @ 1000 cycles | [147] |
|              | g-C\(_{3}\)N\(_{4}\)-rGO | In-situ chemical synthetic approach | 2D sheets | 3002 @ 100 mA/g | 57% | 1525 @ 50 cycles | [108] |
|              | CN-rGO    | Hydrothermal synthesis | 3D Sandwich architecture | 1632 @ 50 mA/g | 41.3% | 970 @ 300 cycles | [130] |
|              | g-C\(_{3}\)N\(_{4}\)@rGO | Hydrothermal reaction | Spongy, porous, and tangled ultrathin sheets | 2731 @ 0.1 C | ~89% | 901 @ 50 cycles | [131] |
|              | SN/CN     | Hydrothermal | Cauliflower-like morphology | 733 @ 0.1 C | 55.4% | ~420 @ 60 cycles | [132] |
|              | SnO\(_{2}\)@C\(_{3}\)N\(_{4}\) | Scalable solid-state reaction | Porous and wrinkled material | 1200 @ 0.1 C | ~50% | 550 @ 100 cycles | [149] |
|              | TiO\(_{2}\)@CNS (TCNS) | Self-assembly approach/hydrothermal reaction | Spherical Core-shell particles | 359 @ 0.1 C | ~90% | 303 @ 125 cycles | [100] |
|              | Zn\(_{2}\)GeO\(_{4}\)/g-C\(_{3}\)N\(_{4}\) | Facile solution approach | Ultrathin nanosheet | 1068 @ 200 mA/g | 58.6% | 1370 @ 140 cycles | [109] |
|              | NiCo\(_{2}\)O\(_{4}\)/g-C\(_{3}\)N\(_{4}\) | Facile ultrasonic treatment | Nanosheet structure | 1367 @ 100 mA/g | 84.5% | 1252 @ 100 cycles | [150] |
|              | Li\(_{4}\)Ti\(_{5}\)O\(_{12}\)/g-C\(_{3}\)N\(_{4}\) | Solvothermal method | Nanoparticles | 173.7 @ 0.5 C | – | 150.8 @ 502 cycles | [133] |
|              | Nitrogen-doped TiO\(_{2}\)/C (NCLTO) | Thermal decomposition | Irregular sized nanoparticles | 189 @ 1 C | ~92% | 122 @ 500 cycles | [109] |
|              | CuO/O-doped g-C\(_{3}\)N\(_{4}\) | Hydrothermal | Nanospheres | 980 @ 100 mA/g | 94.7% | 738 @ 100 cycles | [101] |
|              | 3D N-rich C\(_{3}\)N\(_{4}\)@MoS\(_{2}\) | Hydrothermal | Nanosphere | 2390 @ 0.1 C | ~74% | 857 @ 50 cycles | [154] |
|              | MoS\(_{2}\)/g-C\(_{3}\)N\(_{4}\)@H\(^{+}\)/GO | Sonication/thermal treatment | 2D sheet-like structure | 1728 @ 0.1 mA/g | – | 1450 @ 200 cycles | [103] |
|              | MoS\(_{2}\)/g-C\(_{3}\)N\(_{4}\) | Thermal treatment (calcination) | Spherical particles | 2467 @ 0.05 C | ~41% | 1204 @ 200 cycles | [175] |
|              | WS\(_{2}\)/g-C\(_{3}\)N\(_{4}\) | Solid-state reaction | Nano-sized petal-like sheets | 1933.6 @ 100 mA/g | 63.6% | 622.7 @ 400 cycles | [156] |
|              | SnS\(_{2}\)/CN | Microwave hydrothermal method | Nanoflower | 1465.9 @ 100 mA/g | 47% | 383.8 | [153] |
|              | Co\(_{1−x}\)S@g-C\(_{3}\)N\(_{4}\) | Solvothermal method | Spherical-like | ~1250 @ 0.1 A/g | ~55% | 789.59 @ 210 cycles | [155] |
|              | C\(_{3}\)N\(_{2}\)/MoS\(_{2}\) | Soft- and hard templating methods | Highly ordered mesoporous nanosheet | 271 @ 100 mA/g | 60% | 193 @ 100 cycles | [112] |
| **SIBs**     | huCP/g-C\(_{3}\)N\(_{4}\) | Hydrothermal treatment | Carbon fibers with large pores and hollow structure | 222 @ 0.1 A/g | – | 345 @ 380 cycles | [147] |
6.3 Lithium-Sulfur Batteries

Carbon nitrides have proven to be effective at anchoring LiPSs in Li–S batteries, thereby inhibiting their dissolution in the electrolyte and boosting overall battery performance [134, 135]. Experimental battery studies on CNBMs as sulfur hosts for Li–S battery have confirmed the DFT predictions discussed earlier. For example, Liu et al. reported the application of a 2D graphene-like oxygenated C$_3$N$_4$ via a scalable one-step self-supporting solid-state pyrolysis

| Battery type | Materials | Synthesis | Morphology | Initial capacity (mAh g$^{-1}$) | ICE$^a$ | Reversible capacity (mAh g$^{-1}$) | References |
|--------------|-----------|-----------|------------|---------------------------------|--------|----------------------------------|------------|
| N-doped MoC  | Pyrolysis  | Hollow micro-spheres | 1040 @ 0.16 A/g | – | 410@ 200 cycles | [172] |
| Amorphous carbon nitride (ACN) | Copolymerization/direct carbonization | Uniform rhombic dodecahedral shape | 640 @ 83 mA/g | 67.2% | 175 @ 2000 cycles | [173] |
| N-FLG-800  | Pyrolysis  | 2D lamellar structure of with plenty of wrinkles | 256.7 @ 0.5 A/g | 83.5% | 211.3 @ 2000 cycles | [175] |
| C/g-C$_3$N$_4$ | One-pot calcination | 2D-sheet-like structure | ~200 @ 0.4 A/g | 99% | ~ 160 @ 400 cycles | [128] |
| g-C$_3$N$_4$ film | Chemical vapor deposition (CVD) | 2D thin film | – | 98% | ~6 Ah/g @ 500 cycles | [174] |
| CN/MoS$_2$-600 | Nanotemplating approach | Corrugated cardboard-like morphology | – | 70% | 605 | [98] |
| C$_2$N$_2$/MoS$_2$ | Combined soft- and hard templating methods | Ordered mesoporous nanosheet | 126 @ 100 mA/g | 46% | 54 @ 100 cycles | [112] |
| PIBs 1D/2D C$_3$N$_2$/rGO | Hydrothermal/freeze drying method | 1D-C$_3$N$_2$ rod infused in-between the 2D-sheet | 682.7 @ 0.5 A/g | 56.8% | 557.4 @ 50 cycles | [54] |
| PIBs Co$_3$O$_4$@N–C | Ionic liquid-assisted solvothermal method | Spherical | 1229.2 @ 50 mA/g | 48.2% | 448.7 @ 40 cycles | [176] |
| LIBs Si@rGO/g-C$_3$N$_4$ | Template-free self-assembly and pyrolysis process | Multilayered 3D framework | 1354.8 @ 0.5 C | 70.9% | 799.6 @ 1000 cycles | [159] |
| PrGO-C$_3$N$_4$ | Ball milling | Nubby structure (40–200 nm) | 2423.4 @ 200 mA/g | 99% | 1032.6 @ 600 cycles | [160] |
| rGO/C$_3$N$_4$/SnS$_2$ (GSC6) | Hydrothermal route | 3D porous structure | 1073.1 @ 100 mA/g | 61.1% | 1118.6 @ 100 cycles | [122] |
| rGO/g-C$_3$N$_4$@SnS$_2$ | Solvothermal synthesis | Nano-spheres anchored on hybrid sheet | ~600 @ 800 mA/g | —— | 864.9 @ 1000 cycles | [164] |
| MnO/C$_3$N$_4$/C | Sonication/calcination | 3D Porous sphere | 918.9 @ 0.5 C | 66% | 781.9 @ 86 cycles | [165] |
| SnO$_2$-TiO$_2$-C$_3$N$_4$ | Self-assembly deposition | Microspherical shaped NPs | 853 @ 0.1 C | —— | 114.1 @ 20 cycles | [188] |
| Co@Co$_9$S$_8$/S–N-C | Hydrothermal reaction and pyrolysis method | Porous structure with smooth-faced surfaces | 1033.25 @ 0.2 A/g | 60% | 652.1 @ 610 cycles | [166] |

$^a$ICE Initial coulombic efficiency
process (Fig. 22a). The oxygen-rich functional groups possess strong chemical adsorption toward sulfur atoms; hence, they can inhibit the dissolution of polysulfides in Li–S electrolyte. As a cathode material, the resultant electrode delivered an initial columbic efficiency of 98% (S utilization of 84%) with a reversible capacity of 1407.6 mAh g⁻¹.
Carbon coating for anchoring soluble polysulfides is an effective strategy to improve the conductivity of electrode materials in Li–S batteries [135]. Based on the conclusion of DFT calculations, Liao and co-workers showed that coating g-C3N4 on carbon-fiber mesh can serve as an effective anchoring strategy for LiPSs, and the g-C3N4@CFM electrode delivered 905 mAh g⁻¹ at 0.1 C. Figure 22c shows the polysulfide adsorption mechanism of the composite electrode [67]. Liang et al. deployed polymeric C₃N₄ (p-C₃N₄) as a suitable cathode material to effectively attract LiPSs in common electrolyte solvents (1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME)). DFT calculations proved that interaction between p-C₃N₄ and LiPSs can alter the bonding and spatial configuration of the lithium polysulfides, which in turn tunes their redox kinetics. The results of Li–S battery testing in Fig. 22d showed that the electrode consisting of p-C₃N₄ and rGO (CNG) performed better than its individual constituents, improved stabilization, and kinetics of LiPSs and restrained shuttling effect [57].

Coating of the commercial polypropylene separator with g-C₃N₄ is another strategy to facilitate LiPSs adsorption and effectively suppress polysulfide dissolution [134, 135]. Fan et al. reported the blending of commercial separator with carbon black (g-C-coated) and coating it on commercial glass fiber separator for LiPSs adsorption. The g-C₃N₄-coated separator showed better capacity retention in that after 400 cycles at 0.2 C, it could still deliver 773.2 mAh g⁻¹, while the carbon-coated separator only had 611 mAh g⁻¹ [68]. Chen et al. showed that coating of a transition metal (Fe, Ni, Cu, and Co) coordinated C₃N₄/C commercial separator improved LiPSs adsorption and performance. Experimental Li–S battery testing and DFT calculations concluded that Ni-modified C₃N₄ facilitated the LiPSs adsorption the best; hence, it outperformed the other metal modified C₃N₄/C-coated separators in terms of battery performance (Fig. 23a). A schematic of the polysulfide adsorption mechanism of the metal modified electrode is depicted in Fig. 23b. In terms of CNBCs, Qu et al. reported the use of graphene as a conductive material combined with C₃N₄ to form a g-C₃N₄/GS interlayer for a sulfur filled keijten black cathode for Li–S battery. Due to the electrostatic forces present as a result of π–π bonding between both materials, graphene and g-C₃N₄ heterogeneously intercalated with each in a closely packed fashion to form a laminated channel which inhibited diffusion of LiPSs toward the anode (Fig. 23c). The cathode material delivered a high reversible
capacity (> 1200 mAh g⁻¹) when cycled at 1 C and retained up to 50% of its capacity after 100 cycles. Moreover, from Fig. 23d a stable and consistent charge/discharge profile was maintained even when the electrode was cycled at various current densities [137]. To summarize the discussion on Li–S, CNBMs have been used for several functions in Li–S batteries and they have shown great promise as advanced electrode materials (Table 4). However, the percentage of S loading needs to be increased from an average of 50% to an average of ~ 80% for all CNBCs reports. Also, the binding mechanism and chemical interactions between the CNBMs and the binder need to be further investigated.

### 6.4 Lithium-Oxygen Batteries

As discussed earlier, DFT predictions show that carbon nitrides are promising for Li-O₂ battery because they possess a superior overpotential than most commercial electrodes because molecular level interaction with LiO₂ indicates that they form a Li-N bonding which is dependent on the ratio of N in the material and greatly influences their overpotential [60].

Experimental study of pure and CNBCs in Li-O₂ batteries has been reported in the literature. For instance, Liu et al. showed that g-C₃N₄ due to the lower conduction band (CB) potential of g-C₃N₄ and its low bandgap of ~ 2.7 eV the use of g-C₃N₄ together with an I⁻ ion redox mediator and non-aqueous electrolyte resulted in a drastic decrease in charging voltage [139].

Heteroatom doping (especially single atom doping) is a well-known method of modulating the electronic properties [140, 141]. Zhao et al. reported the application of a Pt-doped g-C₃N₄ (CNHS) for Li-O₂ battery, and the performance and cycle stability of Pt-g-C₃N₄ (Pt-CNHS) exceeded those of pure holey g-C₃N₄. A schematic illustration of the Li-O₂ reaction process is presented in Fig. 24a. Also, Li-O₂ battery analysis (charge/discharge voltage plateau) shows that Pt-CNHS possess a lower overpotential when compared to CNHS with a round trip efficiency of 69% compared to 65% for CNHS [63]. Gao et al. also reported the design of a tungsten carbide-modified N-doped defective carbon (W₉C@NC), and the composite exhibited a significant synergistic interaction (Fig. 24b) and delivered an initial reversible capacity of 10,976 mAh g⁻¹ at a current density
| Battery function | Materials | Synthesis | Morphology | Initial capacity (mAh g⁻¹) | Sulfur loading | Reversible capacity (mAh g⁻¹) | References |
|------------------|-----------|-----------|------------|-----------------------------|---------------|-------------------------------|------------|
| Cathode          | OCN       | One-step self-supporting solid-state pyrolysis (OSSP) | Free-standing 2D sheets | 1407.6 @ C/20 | 56 wt% | 447.3 @ 500 cycles | [177] |
|                  | GCN       | Chemical deposition method | Sheet-like morphology | 1250.1 @ C/0.05 | 70.4 wt% | 578.0 @ 750 cycles | [132] |
|                  | C-NC/GN/g-C₃N₄ | Low-temperature dissolution and high-temperature carbonization | 3D interconnected network ultrathin sheets | ~1270 @ 0.5 C | ~78.1 wt% | ~1130 @ 500 cycles | [178] |
|                  | CN@NSHPC  | Double-solvent approach with controllable pyrolysis | Solid spheroidal nano-particle | 1099 @ 0.1 C | 73% (1.7–2 mg/cm²) | 445 @ 500 cycles | [179] |
|                  | CN/MoS₂   | Hydrothermal method | Laminar nanosheet | 1252 @ 0.5 C | 4 mg/cm² | 680 @ 200 cycles | [184] |
|                  | rGO/g-C₃N₄/CNT | Ethanol-assisted spray-drying method | Microsphere-like structure | 1030 @ 0.2 C | 1.5 mg/cm² | 820 @ 200 cycles | [182] |
|                  | 3D PCN@rGO | Hydrothermal and freeze-drying | Interconnected sheets stacked by several layers | 1240 @ 0.2 C | ~1.5 mg/cm² | 1005 @ 170 cycles | [181] |
| Sulfur host      | C₃N₄@CFM  | In situ coating method | Carbon fiber mesh | 905 @ 0.1 C | 0.6 mg/cm² | 801 @ 100 cycles | [140] |
|                  | C₃N₄/rGO aerogel (CG12) | Hydrothermal, freeze-drying method | Inter-connected thin nanosheets | 1024 @ 0.5 C | 80 wt% (~1 mg/cm²) | 770 @ 100 cycles | [138] |
|                  | MoS₂/g-C₃N₄ | Melt-diffusion method | Multi-layered sheet like structure | 943 @ 1 C | ~1.5 mg/cm² (59.1%) | 569 @ 400 cycles | [102] |
| Facilitate LiPS redox | CNG (Polymeric g-C₃N₄ and rGO) | Simple cyro-drying and annealing process | Intercalated thin sheets | ~1250 @ ~330 mA/g | ~0.6 mg of sulfur on 1.2 mg of CNG or r-GO | over 700 @ 280 cycles | [131] |

| Separator coating for Li–S battery |
|-----------------------------------|
| Battery type | Materials | Synthesis | Morphology | Initial capacity (mAh g⁻¹) | Sulfur loading | Reversible capacity (mAh g⁻¹) | References |
| LiPS-trapping separator | C₃N₄/CNT@PP | Thermal condensation and oxidation etching with simple filtration process | Long-range continuous framework | ~1350 @ 0.1 C | 7.2 mg/cm² | 870.5 @ 100 cycles | [180] |
|                  | g-C₃N₄ | Thermal condensation and oxidation etching | Ultrathin nanosheet | ~1000 | 4 mg/cm² | 829 @ 200 cycles | [183] |
of 100 mAh g\(^{-1}\) with a low overpotential and long cycle life [142].

Luo et al. reported the fabrication of a 3D free-standing graphene@g-C\(_3\)N\(_4\) (G@CN) composite which delivered superior performance (~17,300 mAh g\(^{-1}\) at a discharge voltage plateau of 2.6 V), energy efficiency (71%), and stable cycle life compared to the graphene electrode when applied in Li-O\(_2\) battery (Fig. 24c) [143]. Lou et al. also reported the application of mesoporous boron-doped carbon nitride (m-BCN) as a support for even distribution of ruthenium oxide (RuO\(_2\) nanoparticles). As a Li-O\(_2\) cathode, the RuO\(_2@m\)-BCN composite displayed a superior discharge capacity (2.57 mAh cm\(^{-2}\) = 512 mAh g\(^{-1}\) based on the mass of the composite), smaller overpotential (0.18—discharge and 0.54—recharge), and excellent cycle stability even after 126 cycles. The charge/discharge curve for the composite when cycled at 0.3 mA cm\(^{-2}\) is presented in Fig. 24d [144].

Guo and co-workers reported the synthesis of a ternary composite of Co\(_3\)O\(_4\)-modified Ag/g-C\(_3\)N\(_4\) for Li-O\(_2\). The synergistic interaction between these three constituents produced a composite with superior battery performance and catalytic activity with stable cycle life, high reversible capacity, and round-trip efficiency (Fig. 24e, f) [145].

### 6.5 Lithium-Metal Batteries

Despite the benefits of lithium metal as a preferred anode for high energy batteries, its large-scale application is hampered by two major issues: performance and safety [1, 146, 147]. These two problems are related in that they are both caused by Li dendrite growth. Moreover, other issues such as formation of SEI film [148–150] and non-uniform Li\(^+\) deposition are still of concern. These issues have been tackled in different ways, and because of the surface charges and functional groups of g-C\(_3\)N\(_4\), it has been considered a very useful material for these various solutions. For instance, Luan and co-workers applied O- and N-rich graphene-like g-C\(_3\)N\(_4\) as an effective artificial protective layer for Li-metal anode in half and full LIB and Li–S cells. Pristine g-C\(_3\)N\(_4\) (P-G) and acid-treated g-C\(_3\)N\(_4\) (A-G) were tested for Li-metal, and the A-G-Li exhibited the best wettability, structural stability, and least contact angle of almost zero indicating uniform Li\(^+\) distribution (Fig. 25a). From Fig. 25b, c, A-G-Li delivered the highest reversible capacity and superior capacity
Considering the lithophilic nature of g-C₃N₄, its incredible mechanical strength, and unique morphology, Hu et al. reported the use of porous g-C₃N₄ microspheres as a polymeric electrolyte filler for lithium metal anode. Li dendrite was successfully suppressed in the g-C₃N₄ filler (Fig. 25d). It delivered superior performance and stability to the commercial electrolyte (Fig. 25e, f) [152]. Luo et al. reported the uniform coating of g-C₃N₄ on a 3D Ni foam (Fig. 25g) to design a g-C₃N₄@Ni foam which was applied as a current collector for Li-metal anode. The g-C₃N₄@Ni foam anode delivered superior capacity (Fig. 25h, i) to the pure Ni foam with excellent coulombic efficiency (98%), cycle stability, and capacity retention (72.9% at 1 C after 200 cycles) [153].

### 6.6 Zinc-Air Batteries

The affordability, safety, and high specific energy density of ZABs are some of the reasons for the intensive research into this type of metal-air battery technology. However, the high cost and stability of metal-based catalysts for ZABs application [154, 155] have motivated researchers into designing metal-doped carbon materials [156, 157]. The high nitrogen content of C₃N₄ makes it a viable material for composite design or as a reliable precursor for N-doped carbon synthesis. For instance, Shinde et al. developed a 3D carbon NP sandwiched in phosphorus and sulfur co-doped g-C₃N₄ as a metal-free hybrid cathode material for ZAB. Electrochemical battery testing shows that the hybrid air–cathode operated at a voltage of approximately 1.51 V (Fig. 26a),
delivered a high reversible capacity of 830 mAh g\(^{-1}\) along with a high energy density of 970 Wh kg\(^{-1}\), 198 mW cm\(^{-2}\) power density with excellent stability even after been recharged more than 210 h [158]. The composite was also tested in a tri-electrode ZABs system, and a schematic of the tri-electrode ZABs is shown in Fig. 26b.

Shinde et al. also synthesized 3D phosphorus, sulfur co-doped carbon nitride nanofibers. As a cathode material in a primary zinc-air battery, the hybrid nanofiber architecture operated at a 1.49 circuit voltage (consider Fig. 26c) and delivered a reversible capacity of 698 mAh g\(^{-1}\), power and energy density of 231 mW/cm\(^2\) and 785 Wh/kg, respectively, with great stability even after 240 h of operation [159, 160]. Ma et al. designed a flexible ZAB with bimetallic iron and cobalt (FeCo) sandwiched in N-doped carbon nanotubes (N-CNT). As an air–cathode in ZAB (Fig. 26d), the FeCo/N-CNT composite operated at an open-circuit voltage of 1.25 V, delivered a power density of 97.8 mW/cm\(^2\), and showed excellent stability at 100 mA cm\(^{-2}\) even after 144 cycles [160]. Zhang et al. reported the synthesis of a zinc air electrode using g-C\(_3\)N\(_4\) as a template for N-doped carbon. The Fe–N-C air cathode operated at an open-circuit potential of 1.51 V, delivered a power density of 100 mW cm\(^{-2}\) and was able to power a 3 V blue LED lamp. As seen in Fig. 26e, the air–cathode operated with excellent stability at a current density of 10 mA cm\(^{-2}\) even after 6 h outshining the N–C electrode. The cathode materials were able to light up LED lamps; photographs of the LED lights powered by the cathode materials are presented in Fig. 26f [161].

6.7 Solid-State Batteries

Unlike liquid organic electrolyte systems, the possibility of maximizing electron transfer in a solid-state electrolyte and lithium metal battery system is rather arduous because of the reducing property of Li metal when it interacts with most solid-state electrolytes [162]. This reductive process occurs because of the high reduction potential of solid-state electrolytes against that of Li, for example Li\(_{10}\)GeP\(_2\)S\(_{12}\) (1.71 V), Li\(_{0.52}\)Ti\(_2\)Al\(_{0.3}\)(PO\(_4\))\(_3\) (2.17 V), and Li\(_{0.33}\)La\(_{0.56}\)TiO\(_3\) (1.75) [163, 164]. While the reductive potential of Li\(_{7}\)La\(_{3}\)Zr\(_5\)O\(_{12}\) (0.05 V)
is very far from that of Li, it possesses a lithiophobic surface which means that it will experience poor contact and great surface resistance with lithium metal anode. However, this problem can be solved by meticulously coating its surface with a lithophilic material, thereby making changing its lithiophobicity and making Li$_7$La$_3$Zr$_5$O$_{12}$ lithophilic. Huang et al. used this approach when they reported the in situ formation of a Li$_3$N layer between Li and Li$_7$La$_3$Zr$_5$O$_{12}$ such that Li$^+$ can be effectively conducted and Li dendrite formation suppressed. This Li$^+$ conducting and electron-insulating Li$_3$N layer was created because of the reaction between molten Li and g-C$_3$N$_4$ to form a Li-C$_3$N$_4$ composite such that the role of C$_3$N$_4$ is for interface formation between Li and Li$_7$La$_3$Zr$_5$O$_{12}$ (Fig. 27a, b). The composite delivered a high capacity and stable cycle performance when tested in a solid-state battery (Fig. 27c) and maintained a strong/intimate contact with the Garnet (Fig. 27d) [165]. Lu et al. also reported the application of mesoporous carbon nitride (MCN) and acetylene black (AB) as a support for in situ growth of Pt nanoparticles for an all-solid-state lithium-air battery system. A schematic of the assembled MCN-based electrode solid-state lithium-air battery is presented in Fig. 27e. As a cathode material in all-solid-state Li-air battery, the Pt@MCN electrode synthesized by Lu et al. recorded a significant high round trip efficiency (87%) at a high discharge voltage of ~2.87 V along with a low charge voltage of ~3.3 V (Fig. 27f, g) [166]. Sun et al. reported the application of g-C$_3$N$_4$ as a polymeric filler to improve the coupling effect between the amorphous specie and coordination unit of a solid polymer electrolyte in solid-state
batteries. When applied as an electrolyte with LiFePO4 in an all-solid-state battery, 60 °C an initial reversible capacity of 161 mAh g\(^{-1}\) was delivered, and after 100 cycles, 155 mAh g\(^{-1}\) was still retained (Fig. 27h) [167].

7 Conclusions and Future Perspectives

A comprehensive overview of structure-guided synthesis/fabrication and DFT-guided application of CNBMs for energy storage devices has been discussed in this review. CNBMs, including pure, doped, and CNBCs, exhibit high mechanical strength, excellent structural stability, abundant nitrogen-rich active sites, and surface functionalities, which are important for energy storage devices. Through the DFT-guided approach, CNBMs with superior structural and electronic properties are designed and their electrochemical properties can be modulated for improved performance. Moreover, the structure-guided approach facilitates the synthesis and fabrication of pure/doped C\(_x\)N\(_y\) materials and CNBC’s, respectively. The massive attention into CNBMs for electrochemical energy storage and conversion can be attributed to the combination of these two approaches, and they are paramount to the advancement in this field [140, 168–172].

Despite the significant progress made so far in the study of CNBMs for energy storage devices, there are still unresolved issues; therefore, we provide some perspectives to resolve them. Firstly, the important preconditions must be considered when carrying out a DFT-guided synthesis/design of CNBMs. These prerequisites include the crystal phase and lattice parameter of the material, the interlayer...
distance (if it is a multi-layered material) and the exposed surface facet if it is a single-layered material, the ratio of C/N or other elements and type of nitrogen (pyridinic or graphitic-N) dominant in the structure of the material. These preconditions are critical when designing a material via a DFT-guided approach. Secondly, a clear and concise investigation of the metal ion storage mechanism of CNBMs is essential. This will provide a fundamental/atomic-scale understanding of the electrochemical processes occurring at the surface and carbon matrix of the CNBMs. It will also show which surface functionalities of CNBMs impact their metal ion storage mechanism and how the interfacial interactions can be optimized. Thirdly, in situ characterization of CNBMs is required for a deeper understanding of structural changes, surface functional group transformation, and metal ion storage. For example, through in situ Fourier-transform infrared spectroscopy (FTIR), functional groups such as hydroxyl, amine, and carboxylic groups on the surface of CNBMs can be observed. Phase and oxidation state changes can also be identified through in situ X-ray photoelectron spectroscopy (XPS) analysis. These advanced characterization techniques can provide crucial information that will enlighten researchers on the relationship between the electronic properties and the CNBMs. Lastly, theoretical/computational studies of CNBMs have proven to be fundamental in understanding their electronic properties for energy storage devices; hence, it must remain a crucial part of its future research. However, DFT is limited in studying CNBMs for energy storage device because it involves the use of metal atoms instead of ions, and it cannot effectively account for the effect of electrolyte, SEI layer formation, pulverization of electrode, the surface charge of materials and interfacial interactions. Therefore, improvement of existing DFT tools/functions or the development of new ones that can account for these critical parameters is essential to achieve a closer alignment between DFT studies and experimental testing. On a general note, the study of CNBMs is still at the early stage. Therefore, there is much room for improvement, and resolving these issues will surely propel the research to advance at a rapid pace.

Acknowledgements The authors gratefully acknowledge financial support from the Australia Research Council Discovery Projects (DP160102627 and DP1701048343) of Australia and Shenzhen Peacock Plan of China (KQTD201612915051055), and the 111 Project (D20015) of China Three Gorges University.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by/4.0/.

References

1. Z. Su, J. Liu, M. Li, Y. Zhu, S. Qian et al., Defect engineering in titanium-based oxides for electrochemical energy storage devices. Electrochem. Energ. Rev. 3, 286–343 (2020). https://doi.org/10.1007/s41918-020-00064-5

2. Y. Zhang, G. Shi, J. Qin, S.E. Lowe, S. Zhang, H. Zhao, Y.L. Zhong, Recent progress of direct ink writing of electronic components for advanced wearable devices. ACS Appl. Electron. Mater. 1(9), 1718–1734 (2019). https://doi.org/10.1021/acsaelm.9b00428

3. Z. Wu, J. Xie, Z.J. Xu, S. Zhang, Q. Zhang, Recent progress in metal–organic polymers as promising electrodes for lithium/sodium rechargeable batteries. J. Mater. Chem. A 7(9), 4259–4290 (2019). https://doi.org/10.1039/C8TA11994E

4. C. Wu, X. Tong, Y. Ai, D.-S. Liu, P. Yu, J. Wu, Z.M. Wang, A review: enhanced anodes of Li/Na-ion batteries based on yolk–shell structured nanomaterials. Nano-Micro Lett. 10(3), 40 (2018). https://doi.org/10.1007/s40820-018-0194-4

5. C. Liu, X. Liu, J. Tan, Q. Wang, H. Wen, C. Zhang, Nitrogen-doped graphene by all-solid-state ball-milling graphite with urea as a high-power lithium battery anode. J. Power Sources 342, 157–164 (2017). https://doi.org/10.1016/j.jpowsour.2016.11.110

6. B. He, Y. Wang, Q. Zhai, P. Qiu, G. Dong, X. Liu, Y. Chen, Z. Li, From polymeric carbon nitride to carbon materials: extended application to electrochemical energy conversion and storage. Nanoscale 12(16), 8636–8646 (2020). https://doi.org/10.1039/D0NR01612H

7. Q. Hao, G. Jia, W. Wei, A. Vinu, Y. Wang, H. Arandiyan, B.-J. Ni, Graphitic carbon nitride with different dimensionalities for energy and environmental applications. Nano Res. 13(1), 18–37 (2020). https://doi.org/10.1007/s12274-019-2589-z

8. J. Chen, Z. Mao, L. Zhang, D. Wang, R. Xu, L. Bie, B.D. Fahlman, Nitrogen-deficient graphitic carbon nitride with enhanced performance for lithium ion battery anodes. ACS Nano 11(12), 12650–12657 (2017). https://doi.org/10.1021/acsnano.7b07116
9. H. Pang, Y. Luo, Y. Yan, S. Zheng, H. Xue, Graphitic carbon nitride based materials for electrochemical energy storage. J. Mater. Chem. A 7(3), 901–924 (2018). https://doi.org/10.1039/C8TA08464E
10. B. Zhu, B. Cheng, L. Zhang, J. Yu, Review on dft calculation of s-triazine-based carbon nitride. Carbon Energy 1(1), 32–56 (2019). https://doi.org/10.1002/cey2.1
11. A. Bafekry, S.F. Shayesteh, F.M. Peeters, Two-dimensional carbon nitride (2DCN) nanosheets: tuning of novel electronic and magnetic properties by hydrogenation, atom substitution and defect engineering. J. Appl. Phys. 126(21), 215104 (2019). https://doi.org/10.1063/1.5120525
12. R. Shibuya, T. Kondo, J. Nakamura, Active sites in nitrogen-doped carbon materials for oxygen reduction reaction. Carbon-Based Metal-Free Catalysts 1, 227–249 (2018). https://doi.org/10.1002/9783527811458.vol1-ch8
13. K.S. Lakhi, D.-H. Park, K. Al-Bahily, W. Cha, B. Viswanathan, J.-H. Choy, A. Vinu, Mesoporous carbon nitrides: synthesis, functionalization, and applications. Chem. Soc. Rev. 46(1), 72–101 (2017). https://doi.org/10.1039/C6CS00532B
14. H. Xia, Q. Xu, J. Zhang, Recent progress on two-dimensional nanoflake ensembles for energy storage applications. Nano-Micro Lett. 10(4), 66 (2018). https://doi.org/10.1007/s40820-018-0219-z
15. H. Qiu, Z. Wang, X. Sheng, First-principles prediction of an intrinsic half-metallic graphitic hydrogenated carbon nitride. Phys. Lett. A 377(3–4), 347–350 (2013). https://doi.org/10.1016/j.physleta.2012.11.050
16. T. Hussain, D.J. Searles, M. Hankel, Insights into the trapping mechanism of light metals on C,N-h2d: utilisation as an anode material for metal ion batteries. Carbon 160, 125–132 (2020). https://doi.org/10.1016/j.carbon.2019.12.063
17. Y. Zheng, H. Li, H. Yuan, H. Fan, W. Li, J. Zhang, Understanding the anchoring effect of graphene, BN, C,N and C,N4 monolayers for lithium-polyanions in Li–S batteries. Appl. Surf. Sci. 434, 596–603 (2018). https://doi.org/10.1016/j.apsusc.2017.10.230
18. B. Zhu, L. Zhang, B. Cheng, J. Yu, First-principle calculation study of tri-s-triazine-based g-C3N4: a review. Appl. Cat. B Environ. 224, 983–999 (2018). https://doi.org/10.1016/j.apcatb.2017.11.025
19. Q. Weng, G. Li, X. Feng, K. Nielsch, D. Golberg, O.G. Schmidt, Electronic and optical properties of 2D materials constructed from light atoms. Adv. Mater. 30(46), 1801600 (2018). https://doi.org/10.1002/adma.201801600
20. H.-Z. Wu, L.-M. Liu, S.-J. Zhao, The effect of water on the structural, electronic and photocatalytic properties of graphitic carbon nitride. Phys. Chem. Chem. Phys. 16(7), 3299–3304 (2014). https://doi.org/10.1039/C3CP54333A
21. A. Wang, C. Wang, L. Fu, W. Wong-Ng, Y. Lan, Recent advances of graphitic carbon nitride-based structures and applications in catalyst, sensing, imaging, and leds. Nano-Micro Lett. 9(4), 47 (2017). https://doi.org/10.1007/s40820-017-0148-2
22. J. Zhang, Y. Chen, X. Wang, Two-dimensional covalent carbon nitride nanosheets: synthesis, functionalization, and applications. Energy Environ. Sci. 8(11), 3092–3108 (2015). https://doi.org/10.1039/C5EE01895A
23. J. Mahmood, E.K. Lee, M. Jung, D. Shin, H.-J. Choi et al., Two-dimensional polyamine (C,N) from carbonized organic single crystals in solid state. Proc. Natl. Acad. Sci. 113(27), 7414–7419 (2016). https://doi.org/10.1073/pnas.1605318113
24. L. Li, X. Kong, O. Leenaerts, X. Chen, B. Sanyal, F.M. Peeters, Carbon-rich carbon nitride monolayers with dirac cones: dumbbell C,N. Carbon 118, 285–290 (2017). https://doi.org/10.1016/j.carbon.2017.03.045
25. B. Feng, B. Mortazavi, H. Zhang, H. Shao, K. Xu et al., Tuning thermal transport in C,N monolayers by adding and removing carbon atoms. Phys. Rev. Appl. 10(3), 034046 (2018). https://doi.org/10.1103/PhysRevApplied.10.034046
26. C. Pu, D. Zhou, Y. Li, H. Liu, Z. Chen, Y. Wang, Y. Ma, Two-dimensional C,N global minima: unique structural topologies and nanoelectronic properties. J. Phys. Chem. C 121(5), 2669–2674 (2017). https://doi.org/10.1021/acs.jpcc.6b09960
27. D. Wang, H. Li, L. Zhang, Z. Sun, D. Han, L. Niu, J. Zhao, 2D nitrogen-containing carbon material C,N as potential host material for lithium polysulfides: a first-principles study. Adv. Theory Simul. 2(2), 1800165 (2019). https://doi.org/10.1002/ads.201800165
28. H. Lin, R. Jin, A. Wang, S. Zhu, H. Li, Transition metal embedded C,N with efficient polysulfide immobilization and catalytic oxidation for advanced lithium-sulfur batteries: a first-principles study. Ceram. Int. 45(14), 17996–18002 (2019). https://doi.org/10.1016/j.ceramint.2019.06.018
29. B. Tian, T. Huang, J. Guo, H. Shu, Y. Wang, J. Dai, Performance effects of doping engineering on graphene-like C,N as an anode material for alkali metal ion batteries. Mater. Sci. Semicond. Proc. 109, 104946 (2020). https://doi.org/10.1016/j.mssp.2020.104946
30. M. Xu, H. Wang, S. Sun, H. Li, X. Li, Y. Chen, Y. Ni, First-principles study of metal atoms adsorption on 2D dumbbell C,N. Phys. Status Solidi B 257(1), 1900205 (2020). https://doi.org/10.1002/pssb.201900205
31. W. Cha, I.Y. Kim, J.M. Lee, S. Kim, K. Ramadass, K. Gopalan Kishnan, S. Premkumar, S. Umapathy, A. Vinu, Sulphur-doped mesoporous carbon nitride with an ordered porous structure for sodium-ion batteries. ACS Appl. Mater. Interfaces 11(30), 27192–27199 (2019). https://doi.org/10.1021/acsami.9b07657
32. M. Molaei, S.M. Mousavi-Khoshdel, M. Ghiasi, Exploring the effect of phosphorus doping on the utility of g-C3N4 as an electrode material in Na-ion batteries using DFT method. J. Mol. Model. 25(8), 256 (2019). https://doi.org/10.1007/s00894-019-4109-1
33. M.-S. Balogun, Z. Wu, Y. Luo, W. Qiu, X. Fan, B. Long, M. Huang, P. Liu, Y. Tong, High power density nitridated hematite (α-Fe2O3) nanorods as anode for high-performance flexible lithium ion batteries. J. Power Sources 308, 7–17 (2016). https://doi.org/10.1016/j.jpowsour.2016.01.043
34. S. Kim, M. Hankel, W. Cha, G. Singh, J.M. Lee, I.Y. Kim, A. Vinu, Theoretical and experimental investigations of...
mesoporous C3N4/MoS2 hybrid for lithium and sodium ion batteries. Nano Energy 72, 104702 (2020). https://doi.org/10.1016/j.nanoen.2020.104702
35. X. Wu, F. Kang, W. Duan, J. Li, Density functional theory calculations: a powerful tool to simulate and design high-performance energy storage and conversion materials. Prog. Nat. Sci. 29(3), 247–255 (2019). https://doi.org/10.1016/j.pnsc.2019.04.003
36. M. Li, T. Gould, Z. Su, S. Li, F. Pan, S. Zhang, Electrochromic properties of Li2TeO3: from visible to infrared spectrum. Appl. Phys. Lett. 115(7), 073902 (2019). https://doi.org/10.1063/1.5099330
37. Y. Wang, Z. Jiao, S. Ma, Y. Guo, Probing C3N/graphene heterostructures as anode materials for Li-ion batteries. J. Power Sources 413, 117–124 (2019). https://doi.org/10.1016/j.jpowsour.2018.12.031
38. Y. Ding, B. Xiao, J. Li, Q. Deng, Y. Xu, H. Wang, D. Rao, Improved transport properties and novel Li diffusion dynamics in van der waals C3N/graphene heterostructure as anode materials for lithium-ion batteries: a first-principles investigation. J. Phys. Chem. C 123(6), 3353–3367 (2019). https://doi.org/10.1021/acs.jpcc.8b12379
39. D. Adekoya, S. Zhang, M. Hankel, 1D/2D C3N4/graphene composite as a preferred anode material for lithium ion batteries: importance of heterostructure design via DFT computation. ACS Appl. Mater. Interfaces 12(23), 25875–25883 (2020). https://doi.org/10.1021/acsami.0c04900
40. Q. Liu, B. Xiao, J. Cheng, Y.-C. Li, Q. Li et al., Carbon excess C3N: a potential candidate as Li-ion battery material. ACS Appl. Mater. Interfaces 10(43), 37135–37141 (2018). https://doi.org/10.1021/acsami.8b14183
41. A. Jain, Y. Shin, K.A. Persson, Computational predictions of energy materials using density functional theory. Nat. Rev. Mater. 1(1), 15004 (2016). https://doi.org/10.1038/natrevmats.2015.4
42. M. Wu, Q. Wang, Q. Sun, P. Jena, Functionalized graphitic carbon nitride for efficient energy storage. J. Phys. Chem. C 117(12), 6055–6059 (2013). https://doi.org/10.1021/jp311972f
43. Y. Mao, H. Duan, B. Xu, L. Zhang, Y. Hu et al., Lithium storage in nitrogen-rich mesoporous carbon materials. Energy Environ. Sci. 5(7), 7950–7955 (2012). https://doi.org/10.1039/c2ee21817h
44. G.M. Veith, L. Baggetto, L.A. Adamczyk, B. Guo, S.S. Brown et al., Electrochemical and solid-state lithiation of graphitic C3N4. Chem. Mater. 25(3), 503–508 (2013). https://doi.org/10.1021/cm303870x
45. M. Hankel, D. Ye, L. Wang, D.J. Searles, Lithium and sodium storage on graphitic carbon nitride. J. Phys. Chem. C 119(38), 21921–21927 (2015). https://doi.org/10.1021/acs.jpcc.5b07572
46. D. Adekoya, X. Gu, M. Rudge, W. Wen, C. Lai, M. Hinkel, S. Zhang, Carbon nitride nanofibres with exceptional lithium storage capacity: from theoretical prediction to experimental implementation. Adv. Funct. Mater. 28(50), 1803972 (2018). https://doi.org/10.1002/adfm.201803972
47. H. Pan, Graphitic carbon nitride nanotubes as Li-ion battery materials: a first-principles study. J. Phys. Chem. C 118(18), 9318–9323 (2014). https://doi.org/10.1021/jp412272
48. M. Hankel, D.J. Searles, Lithium storage on carbon nitride, graphenylene and inorganic graphenylene. J. Phys. Chem. C 18(21), 14205–14215 (2016). https://doi.org/10.1039/C5CP07356A
49. D. Wu, B. Yang, H. Chen, E. Ruckenstein, Nitrogenated holey graphene C2N monolayer anodes for lithium- and sodium-ion batteries with high performance. Energy Storage Mater. 16, 574–580 (2019). https://doi.org/10.1016/j.ensm.2018.09.001
50. J. Xu, J. Mahmood, Y. Dou, S. Dou, F. Li, L. Dai, J.B. Baek, 2D frameworks of C2N and C3N as new anode materials for lithium-ion batteries. Adv. Mater. 29(34), 1702007 (2017). https://doi.org/10.1002/adma.201702007
51. G. Guo, R. Wang, S. Luo, B. Ming, C. Wang et al., Metallic two-dimensional C2N allotropes with electron and ion channels for high-performance Li-ion battery anode materials. Appl. Surf. Sci. 518, 146254 (2020). https://doi.org/10.1016/j.apsusc.2020.146254
52. C. Yang, X. Zhang, J. Ma, B. Shi, H. Zhang et al., Ultra-high capacity of monolayer dumbbell C2N as a promising anode material for lithium-ion battery. J. Electrochem. Soc. 167(2), 020538 (2020). https://doi.org/10.1149/1945-7111/ab6bbd
53. G.M. Weng, Y. Xie, H. Wang, C. Karpovich, J. Lipton et al., A promising carbon/g-C3N4 composite negative electrode for a long-life sodium-ion battery. Angew. Chem. Int. Ed. 131(39), 13865–13871 (2019). https://doi.org/10.1002/ange.201905803
54. D. Adekoya, M. Li, M. Hankel, C. Lai, M.-S. Balogun, Y. Tong, S. Zhang, Design of a 1D/2D C3N4/rGO composite as an anode material for stable and effective potassium storage. Energy Storage Mater. 25, 495–501 (2019). https://doi.org/10.1016/j.ensm.2019.09.033
55. P. Bhauriyal, A. Mahata, B. Pathak, Graphene-like carbon–nitride monolayer: a potential anode material for Na-and K-ion batteries. J. Phys. Chem. C 122(5), 2481–2489 (2018). https://doi.org/10.1021/acs.jpcc.7b09433
56. T. Li, C. He, W. Zhang, A novel porous C4N4 monolayer as a potential anchoring material for lithium–sulfur battery design. J. Mater. Chem. A 7(8), 4134–4144 (2019). https://doi.org/10.1039/C8TA10933H
57. J. Liang, L. Yin, X. Tang, H. Yang, W. Yan et al., Kinetically enhanced electrochemical redox of polysulfides on polymeric carbon nitrides for improved lithium–sulfur batteries. ACS Appl. Mater. Interfaces 8(38), 25193–25201 (2016). https://doi.org/10.1021/acsami.6b05647
58. Z. Meng, Y. Xie, T. Cai, Z. Sun, K. Jiang, W.-Q. Han, Graphene-like g-C3N4 nanosheets/sulfur as cathode for lithium–sulfur battery. Electrochim. Acta 210, 829–836 (2016). https://doi.org/10.1016/j.electacta.2016.06.032
59. S.S. Shinde, C.H. Lee, J.-Y. Yu, D.-H. Kim, S.U. Lee, J.-H. Lee, Hierarchically designed 3D holey C2N aerogels as bifunctional oxygen electrodes for flexible and rechargeable...
62. G.-M. Weng, Y. Xie, H. Wang, C. Karpovich, J. Lipton et al., C3N/blue phosphorene heterostructure as a potential anode material of lithium-ion batteries. Appl. Surf. Sci. 510, 145324 (2020). https://doi.org/10.1016/j.apsusc.2020.145324

63. W. Zhao, J. Wang, R. Yin, B. Li, X. Huang, L. Zhao, L. Qian, Single-atom pt supported on holey ultrathin g-C3N4 nanosheets as efficient catalyst for Li-O2 batteries. J. Mater. Chem. A 7(5), 2106–2113 (2019). https://doi.org/10.1039/C8TA0972A

64. H. Lin, R. Jin, S. Zhu, Y. Huang, C3N/blue phosphorene heterostructure as a high rate-capacity and stable anode material for lithium ion batteries: insight from first principles calculations. Appl. Surf. Sci. 505, 144518 (2020). https://doi.org/10.1016/j.apsusc.2019.144518

65. K. Liao, P. Mao, N. Li, M. Han, J. Yi, Y. Sun, H. Zhou, Stabilization of polysulphides via lithium bonds for Li-S batteries. J. Mater. Chem. A 4(15), 5406–5409 (2016). https://doi.org/10.1039/C6TA00054A

66. N. Feng, F. Liu, M. Huang, A. Zheng, Q. Wang et al., Unravelling the efficient photocatalytic activity of boron-induced Ti3+ species in the surface layer of TiO2. Sci. Rep. 6, 34765 (2016). https://doi.org/10.1038/srep34765

67. M. Chen, X. Zhao, Y. Li, P. Zeng, H. Liu et al., Kinetically elevated redox conversion of polysulphides of lithium-sulfur battery using a separator modified with transition metals coordinated g-C3N4 with carbon-conjugated. Chem. Eng. J. 385, 123905 (2020). https://doi.org/10.1016/j.cej.2019.123905

68. B. Xiao, T. Rojo, X. Li, Hard carbon as sodium-ion battery anodes: progress and challenges. ChemSusChem 12(1), 133–144 (2019). https://doi.org/10.1002/cssc.201801879

69. V. Gabaudan, L. Stievano, L. Monconduit, R. Berthelot, Snapshot on negative electrode materials for potassium-ion batteries. Front. Energy Res. 7, 46 (2019). https://doi.org/10.3389/fenrg.2019.00046

70. A.M. Garay-Tapia, A.H. Romero, V. Barone, Lithium adsorption on graphene: from isolated adatoms to metallic sheets. J. Chem. Theory Comput. 8(3), 1064–1071 (2012). https://doi.org/10.1021/ct300042p

71. S. Ullah, P.A. Denis, F. Sato, Unusual enhancement of the adsorption energies of sodium and potassium in sulfur – nitrogen and silicon – boron codoped graphene. ACS Omega 3(11), 15821–15828 (2018). https://doi.org/10.1021/acsomega.8b02500

72. A. Lugo-Solis, I. Vasiliev, Ab initio study of K adsorption on graphene and carbon nanotubes: role of long-range ionic forces. Phys. Rev. B 76(23), 235431 (2007). https://doi.org/10.1103/PhysRevB.76.235431

73. T. Liu, Z. Jin, D.-X. Liu, C. Du, L. Wang, H. Lin, Y. Li, A density functional theory study of high-performance pre-lithiated MS2 (M = Mo, W, V) monolayers as the anode material of lithium ion batteries. Sci. Rep. 10(1), 6897 (2020). https://doi.org/10.1038/s41598-020-63743-9

74. X. Sun, Z. Wang, Y.Q. Fu, Adsorption and diffusion of sodium on graphene with grain boundaries. Carbon 116, 415–421 (2017). https://doi.org/10.1016/j.carbon.2017.01.024

75. J. Yang, Y. Yuan, G. Chen, First-principles study of potassium adsorption and diffusion on graphene. Mol. Phys. 118(1), 1–7 (2019). https://doi.org/10.1080/00268976.2019.1581291

76. A. Eftekhar, Low voltage anode materials for lithium-ion batteries. Energy Storage Mater. 7, 157–180 (2017). https://doi.org/10.1016/j.ensm.2017.01.009

77. J.-H. Kim, D.K. Kim, Conversion-alloying anode materials for na-ion batteries: recent progress, challenges, and perspective for the future. J. Korean Ceram. Soc. 55(4), 307–324 (2018). https://doi.org/10.4191/jkers.2018.55.4.07

78. Z. Jian, W. Luo, X. Ji, Carbon electrodes for K-ion batteries. J. Am. Chem. Soc. 139(12), 7159–7329 (2016). https://doi.org/10.1021/jacs.6b00075

79. H. Yin, Q. Guo, D. He, J. Li, S. Sun, Structural characterization and electrochemical performance of macroporous graphite-like C3N4 prepared by the wurtz reaction and heat treatment. RSC Adv. 7(69), 44001–44008 (2017). https://doi.org/10.1039/C7RA07707F

80. J. Zhu, P. Xiao, H. Li, S.A.C. Carabineiro, Graphitic carbon nitride: synthesis, properties, and applications in catalysis. ACS Appl. Mater. Interfaces. 6(19), 16449–16465 (2014). https://doi.org/10.1021/am502925j

81. Y. Gong, M. Li, Y. Wang, Carbon nitride in energy conversion and storage: recent advances and future prospects. ChemSusChem 8(6), 931–946 (2015). https://doi.org/10.1002/cssc.201403287

82. W.-J. Ong, L.-L. Tan, Y.H. Ng, S.-T. Yong, S.-P. Chai, Graphitic carbon nitride (g-C3N4)-based photocatalysts for artificial photosynthesis and environmental remediation: are we a step closer to achieving sustainability? Chem. Rev. 116(12), 7159–7329 (2016). https://doi.org/10.1021/acs.chemrev.6b00075

83. X. Dong, F. Cheng, Recent development in exfoliated two-dimensional g-C3N4 nanosheets for photocatalytic
applications. J. Mater. Chem. A 3(47), 23642–23652 (2015). https://doi.org/10.1039/C5TA07374J
86. Q. Guo, Q. Yang, C. Yi, L. Zhu, Y. Xie, Synthesis of carbon nitrides with graphite-like or onion-like lamellar structures via a solvent-free route at low temperatures. Carbon 43(7), 1386–1391 (2005). https://doi.org/10.1016/j.carbon.2005.01.005
87. J. Li, C. Cao, J. Hao, H. Qiu, Y. Xu, H. Zhu, Self-assembled one-dimensional carbon nitride architectures. Diam. Relat. Mater. 15(10), 1593–1600 (2006). https://doi.org/10.1016/j.diamond.2006.01.013
88. P. Niu, L. Zhang, G. Liu, H.M. Cheng, Graphene like carbon nitride nanosheets for improved photocatalytic activities. Adv. Funct. Mater. 22(22), 4763–4770 (2012). https://doi.org/10.1002/adfm.201200922
89. J. Mahmood, E.K. Lee, M. Jung, D. Shin, I.-Y. Jeon et al., Nitrogenated holey two-dimensional structures. Nat. Commun. 6(1), 6486 (2015). https://doi.org/10.1038/ncomms7486
90. X. Zhang, X. Xie, H. Wang, J. Zhang, B. Pan, Y. Xie, Enhanced photoresponsive ultrathin graphitic-phase C2N4 nanosheets for bioimaging. J. Am. Chem. Soc. 135(1), 18–21 (2013). https://doi.org/10.1021/ja308249k
91. K. Schwinghammer, M.B. Mesch, V. Duppel, C. Ziegler, J. Senker, B.V. Lotsch, Crystalline carbon nitride nanosheets for improved visible-light hydrogen evolution. J. Am. Chem. Soc. 136(5), 1730–1733 (2014). https://doi.org/10.1021/ja411321s
92. Y. Yin, J. Han, X. Zhang, Y. Zhang, J. Zhou et al., Facile synthesis of few-layer-thick carbon nitride nanosheets by liquid ammonia-assisted lithiation method and their photocatalytic redox properties. RSC Adv. 4(62), 32690–32697 (2014). https://doi.org/10.1039/C4RA06036A
93. Y. Fukasawa, K. Takanabe, A. Shimojima, M. Antonietti, K. Domen, T. Okubo, Synthesis of ordered porous graphitic-C2N4 and regularly arranged Ta2N5 nanoparticles by using self-assembled silica nanospheres as a primary template. Chem. Asian J. 6(1), 103–109 (2011). https://doi.org/10.1002/asia.201000523
94. Y.-S. Jun, W.H. Hong, M. Antonietti, A. Thomas, Mesoporous, 2D hexagonal carbon nitride and titanium nitride/carbon composites. Adv. Mater. 21(42), 4270–4274 (2009). https://doi.org/10.1002/adma.200803500
95. S.S. Park, S.-W. Chu, C. Xue, D. Zhao, C.-S. Ha, Facile synthesis of mesoporous carbon nitrides using the incipient wetness method and the application as hydrogen adsorbent. J. Mater. Chem. 21(29), 10801–10807 (2011). https://doi.org/10.1039/C1JM10849B
96. X.-H. Li, J. Zhang, X. Chen, A. Fischer, A. Thomas, M. Antonietti, X. Wang, Condensed graphitic carbon nitride nanorods by nanoconfinement: promotion of crystallinity on photocatalytic conversion. Chem. Mater. 23(19), 4344–4348 (2011). https://doi.org/10.1021/cm201688v
97. J. Liu, Y. Liu, N. Liu, Y. Han, X. Zhang et al., Metal-free efficient photocatalyst for stable visible water splitting via a two-electron pathway. Science 347(6225), 970–974 (2015). https://doi.org/10.1126/science.aaa3145
98. S. Kim, W. Cha, K. Ramadass, G. Singh, I.Y. Kim, A. Vinu, Single-step synthesis of mesoporous carbon nitride/molybdenum sulfide nanohybrids for high-performance sodium-ion batteries. Chem. Asian J. 15(12), 1863–1868 (2020). https://doi.org/10.1002/asia.202000349
99. J.P. Parakonwitsch, J. Zhang, D. Su, A. Thomas, M. Antonietti, Ionic liquids as precursors for nitrogen-doped graphitic carbon. Adv. Mater. 22(1), 87–92 (2010). https://doi.org/10.1002/adma.200900965
100. Z. Lin, X. Wang, Ionic liquid promoted synthesis of conjugated carbon nitride photocatalysts from urea. Chemsuschem 7(6), 1547–1550 (2014). https://doi.org/10.1002/cssc.201400016
101. Y. Wang, X. Wang, M. Antonietti, Y. Zhang, Facile one-pot synthesis of nanoporous carbon nitride solids by using soft templates. Chemsuschem 3(4), 435–439 (2010). https://doi.org/10.1002/cssc.200900284
102. H. Yan, Soft-templating synthesis of mesoporous graphitic carbon nitride with enhanced photocatalytic H2 evolution under visible light. Chem. Commun. 48(28), 3430–3432 (2012). https://doi.org/10.1039/C2CC00001F
103. Y. Wang, J. Zhang, X. Wang, M. Antonietti, H. Li, Boron- and fluorine-containing mesoporous carbon nitride polymers: metal-free catalysts for cyclohexane oxidation. Angew. Chem. Int. Ed. 49(19), 3356–3359 (2010). https://doi.org/10.1002/anie.201000120
104. Z. Yang, Y. Zhang, Z. Schnepp, Soft and hard templating of graphitic carbon nitride. J. Mater. Chem. A 3(27), 14081–14092 (2015). https://doi.org/10.1039/C5TA02156A
105. T. Kesavan, T. Partheeban, M. Vivekanantha, N. Prabu, M. Kundu et al., Design of P-doped mesoporous carbon nitrides as high-performance anode materials for Li-ion battery. ACS Appl. Mater. Interfaces. 12(21), 24007–24018 (2020). https://doi.org/10.1021/acsami.0c05123
106. Y. Dou, L. Zhang, X. Xu, Z. Sun, T. Liao, S.X. Dou, Atomically thin non-layered nanomaterials for energy storage and conversion. Chem. Soc. Rev. 46(23), 7338–7373 (2017). https://doi.org/10.1039/C7CS00418D
107. Z. Wang, H. Gao, Q. Zhang, Y. Liu, J. Chen, Z. Guo, Recent advances in 3D graphene architectures and their composites for energy storage applications. Small 15(3), 1803858 (2019). https://doi.org/10.1002/smll.201803858
108. D. Adekoya, H. Chen, H.Y. Hoh, T. Gould, M.-S.J.T. Balogun, C. Lai, H. Zhao, S. Zhang, Hierarchical Co9S8@N-doped carbon composite as an advanced anode material for ultrastable potassium storage. ACS Nano 14(4), 5027–5035 (2020). https://doi.org/10.1021/acsnano.0c01395
109. W.-J. Ong, L.-L. Tan, S.-P. Chai, S.-T. Yong, A.R. Mohamed, Surface charge modification via protonation of graphitic carbon nitride (g-C3N4) for electrostatic self-assembly construction of 2D/2D reduced graphene oxide (rGO)/g-C3N4 nanostructures toward enhanced photocatalytic reduction of carbon dioxide to methane. Nano Energy 13, 757–770 (2015). https://doi.org/10.1016/j.nanoen.2015.03.014
110. Y. Fu, J. Zhu, C. Hu, X. Wu, X. Wang, Covalently coupled hybrid of graphitic carbon nitride with reduced graphene
oxide as a superior performance lithium-ion battery anode. Nanoenergy 8, 157–164 (2014). https://doi.org/10.1016/j.nanoen.2014.06.003

112. M.S.A.S. Shah, A.R. Park, A. Rauf, S.H. Hong, Y. Choi et al., Highly interdigitated and porous architected ternary composite of SnS2, g-C3N4, and reduced graphene oxide (RGO) as high performance lithium ion battery anodes. RSC Adv. 7(6), 3125–3135 (2017). https://doi.org/10.1039/C6RA25886G

113. K. Liu, J. Man, J. Cui, H. Zhang, T. Li, J. Yang, Z. Wen, Fabrication of rGO/g-C3N4/SnS2 and its rate-performance enhancement. Chem. Phys. Lett. 746, 137296 (2020). https://doi.org/10.1016/j.cplett.2020.137296

114. W. Shi, T. Wu, X. Song, J. Liu, L. Zhao, P. Zhang, L. Gao, Active Fe3O4 nanoparticles encapsulated in porous gC3N4/graphene sandwich-type nanosheets as a superior anode for high-performance lithium-ion batteries. J. Mater. Chem. A 4(27), 10666–10672 (2016). https://doi.org/10.1039/C6TA03533G

115. J.Y. Hwang, S.T. Myung, Y.K. Sun, Recent progress in rechargeable potassium batteries. Adv. Funct. Mater. 28(43), 1802938 (2018). https://doi.org/10.1002/adfm.201802938

116. K. Song, C. Liu, L. Mi, S. Chou, W. Chen, C. Shen, Recent progress on the alloy-based anode for sodium-ion batteries and potassium-ion batteries. Small 11, 1903194 (2019). https://doi.org/10.1002/smll.201903194

117. Z. Ali, T. Zhang, M. Asif, L. Zhao, Y. Yu, Y. Hou, Transition metal chalcogenide anodes for sodium storage. Mater. Today 3(35), 131–167 (2020). https://doi.org/10.1016/j.mattod.2019.11.008

118. J. Liu, Y. Zhang, L. Zhang, F. Xie, A. Vasileff, S.-Z. Qiao, Graphitic carbon nitride (g-C3N4)-derived n-rich graphene with tuneable interlayer distance as a high-rate anode for sodium-ion batteries. Adv. Mater. 31(24), 1901261 (2019). https://doi.org/10.1002/adma.201901261

119. L. Chen, R. Yan, M. Oschatz, L. Jiang, M. Antonietti, K. Xiao, Ultrathin 2D graphitic carbon nitride on metal films: underpotential sodium deposition in adlayers for sodium-ion batteries. Angew. Chem. Int. Ed. 59(23), 9067 (2020). https://doi.org/10.1002/anie.202000314

120. C.M. Subramaniyam, K.A. Deshmukh, Z. Tai, N. Mahmood, A.D. Deshmukh et al., 2D layered graphitic carbon nitride sandwiched with reduced graphene oxide as nanorchitectured anode for highly stable lithium-ion battery. Electrochim. Acta 237(20), 69–77 (2017). https://doi.org/10.1016/j.electacta.2017.03.194

121. C. Senthil, T. Kesavan, A. Bhaumik, M. Sasidharan, N-rich graphitic carbon nitride functionalized graphene oxide nanosheet hybrid as anode for high performance lithium-ion batteries. Mater. Res. Express 5(1), 016307 (2018). https://doi.org/10.1088/2053-1591/aaa6b9

122. V. Vo, X.D. Nguyen Thi, Y.-J. Jin, G. Ly Thi, T.T. Nguyen, T.Q. Duong, S.-J. Kim, SnO2 nanosheets/g-C3N4 composite with improved lithium storage capabilities. Chem. Phys. Lett. 674, 42–47 (2017). https://doi.org/10.1016/j.cplett.2017.02.057

123. K. Liu, J. Man, J. Cui, H. Zhang, T. Li, J. Yang, Z. Wen, J. Sun, Li4Ti5O12/g-C3N4 composite with an improved lithium...
storage capability. Mater. Lett. 234, 117–120 (2019). https://doi.org/10.1016/j.matlet.2018.09.083

134. Y. Wang, X. Huang, S. Zhang, Y. Hou, Sulfur hosts against the shuttle effect. Small Methods 2(6), 1700345 (2018). https://doi.org/10.1002/smtd.201700345

135. L. Hencz, H. Chen, H.Y. Ling, Y. Wang, C. Lai, H. Zhao, S. Zhang, Housing sulfur in polymer composite frameworks for Li–S batteries. Nano-Micro Lett. 11(1), 17 (2019). https://doi.org/10.1007/s40820-019-0249-1

136. J. Liu, W. Li, L. Duan, X. Li, L. Ji et al., A graphene-like oxygenated carbon nitride material for improved cycle-life lithium/sulfur batteries. Nano Lett. 15(8), 5137–5142 (2015). https://doi.org/10.1021/acs.nanolett.5b01919

137. L. Qu, P. Liu, Y. Yi, T. Wang, P. Yang et al., Enhanced cycling performance for lithium–sulfur batteries by a laminated 2D g-C3N4/graphene cathode interlayer. Chemsuschem 12(1), 213–223 (2019). https://doi.org/10.1002/cssc.201802449

138. J. Song, S. Feng, C. Zhu, J.-I. Lee, S. Fu, P. Dong, M.-K. Song, Y. Lin, Tuning the structure and composition of graphite-phase polymeric carbon nitride/reduced graphene oxide composites towards enhanced lithium-sulfur batteries performance. Electrochim. Acta 248, 541–546 (2017). https://doi.org/10.1016/j.electacta.2017.07.149

139. Y. Liu, N. Li, S. Wu, K. Liao, K. Zhu, J. Yi, H. Zhou, Reducing the charging voltage of a Li−O2 battery to 1.9 V by incorporating a photocatalyst. Energy Environ. Sci. 8(9), 2664–2667 (2015). https://doi.org/10.1039/C5EE01958C

140. Z. Zhao, Y. Sun, F. Dong, Graphitic carbon nitride based nanocomposites: a review. Nanoscale 7(1), 15–37 (2015). https://doi.org/10.1039/c4nr03008g

141. E. Pomerantseva, Y. Gogotsi, Two-dimensional heterostructures for energy storage. Nat. Energy 2(7), 17089 (2017). https://doi.org/10.1038/nenergy.2017.89

142. R. Gao, Y. Zhou, X. Liu, J. Wang, N-doped defective carbon layer encapsulated W,C as a multifunctional cathode catalyst for high performance Li−O2 battery. Electrochim. Acta 245, 430–437 (2017). https://doi.org/10.1016/j.electacta.2017.05.177

143. W.B. Luo, S.L. Chou, J.Z. Wang, Y.C. Zhai, H.K. Liu, A metal-free, free-standing, macroporous graphene@ g-C3N4 composite air electrode for high-energy lithium oxygen batteries. Small 11(23), 2817–2824 (2015). https://doi.org/10.1002/smll.201403535

144. P. Lou, Z. Cui, X. Guo, Achieving highly stable Li−O2 battery operation by designing a carbon nitride-based cathode towards a stable reaction interface. J. Mater. Chem. A 5(34), 18207–18213 (2017). https://doi.org/10.1039/C7TA05009G

145. Q. Guo, C. Zhang, C. Zhang, S. Xin, P. Zhang et al., Co3O4 modified Ag/g-C3N4 composite as a bifunctional cathode for lithium-oxygen battery. J. Energy Chem. 41, 185–193 (2020). https://doi.org/10.1016/j.jenergy.2019.05.018

146. H. Cha, J. Kim, Y. Lee, J. Cho, M. Park, Issues and challenges facing flexible lithium-ion batteries for practical application. Small 14(43), 1702989 (2018). https://doi.org/10.1002/smll.201702989

147. T. Xiong, H. Su, F. Yang, Q. Tan, P.B.S. Appadurai et al., Harmonizing self-supportive VN/MoS2 pseudocapacitance core-shell electrodes for boosting the areal capacity of lithium storage. Mater. Today Energy 17, 100461 (2020). https://doi.org/10.1016/j.mtener.2020.100461

148. X.-Y. Yue, X.-L. Li, W.-W. Wang, D. Chen, Q.-Q. Qiu et al., Wettable carbon felt framework for high loading Li-metal composite anode. Nano Energy 60, 257–266 (2019). https://doi.org/10.1016/j.nanoen.2019.03.057

149. K. Shen, Z. Wang, X. Bi, Y. Ying, D. Zhang et al., Magnetic field–suppressed lithium dendrite growth for stable lithium-metal batteries. Adv. Energy Mater. 9(20), 1900260 (2019). https://doi.org/10.1002/aenm.201900260

150. X.-Y. Yue, W.-W. Wang, Q.-C. Wang, J.-K. Meng, X.-X. Wang et al., Cuprite-coated cu foam skeleton host enabling lateral growth of lithium dendrites for advanced Li metal batteries. Energy Storage Mater. 21, 180–189 (2019). https://doi.org/10.1016/j.ensm.2018.12.007

151. X. Luan, C. Wang, C. Wang, X. Gu, J. Yang, Y. Qian, Stable lithium deposition enabled by an acid-treated g-C3N4 interface layer for a lithium metal anode. ACS Appl. Mater. Interfaces 12(9), 11265–11272 (2020). https://doi.org/10.1021/acsami.9b23520

152. J. Hu, J. Tian, C. Li, Nanostructured carbon nitride polymer-reinforced electrolyte to enable dendrite-suppressed lithium metal batteries. ACS Appl. Mater. Interfaces 9(13), 11615–11625 (2017). https://doi.org/10.1021/acsami.7b00478

153. Z. Lu, Q. Liang, B. Wang, Y. Tao, Y. Zhao et al., Graphitic carbon nitride induced micro–electric field for dendrite-free lithium metal anodes. Adv. Energy Mater. 9(7), 1803186 (2019). https://doi.org/10.1002/aenm.201803186

154. D. Jiao, Z. Ma, J. Li, Y. Han, J. Mao, T. Ling, S. Qiao, Test factors affecting the performance of zinc–air battery. J. Energy Chem. 44, 1–7 (2020). https://doi.org/10.1016/j.jenergy.2019.09.008

155. F. Ran, S. Chen, Advanced Nanomaterials for Electrochemical-based Energy Conversion and Storage (Elsevier, Amsterdam, 2019), p. 416

156. Y.-J. Li, L. Cui, P.-F. Da, K.-W. Qiu et al., Multiscale structural engineering of ni-doped co nanosheets for zinc–air batteries with high power density. Adv. Mater. 30(46), 1804653 (2018). https://doi.org/10.1002/adma.201804653

157. H.-S. Lu, H. Zhang, X. Zhang, N. Sun, X. Zhu, H. Zhao, G. Wang, Transformation of carbon-encapsulated metallic Co into ultratine Co/CoO nanoparticles exposed on N-doped graphitic carbon for high-performance rechargeable zinc-air battery. Appl. Surf. Sci. 448, 369–379 (2018). https://doi.org/10.1016/j.apsusc.2018.04.146

158. S.S. Shinde, C.-H. Lee, A. Sami, D.-H. Kim, S.-U. Lee, J.-H. Lee, Scalable 3D carbon nitride sponge as an efficient metal-free bifunctional oxygen electrocatalyst for rechargeable Zn–air batteries, ACS Nano 11(1), 347–357 (2017). https://doi.org/10.1021/acsnano.6b05914
159. S.S. Shinde, J.-Y. Yu, J.-W. Song, Y.-H. Nam, D.-H. Kim, J.-H. Lee, Highly active and durable carbon nitride fibers as metal-free bifunctional oxygen electrodes for flexible Zn-air batteries. Nanoscale Horiz. 2(6), 333–341 (2017). https://doi.org/10.1039/C7NH00058H

160. C.-Y. Su, H. Cheng, W. Li, Z.-Q. Liu, N. Li et al., Atomic modulation of FeCo–nitrogen–carbon bifunctional oxygen electrodes for rechargeable and flexible all-solid-state zinc–air battery. Adv. Energy Mater. 7(13), 1602420 (2017). https://doi.org/10.1002/aenm.201602420

161. L. Zhang, J. Xiong, Y.-H. Qin, C.-W. Wang, Porous N-C catalyst synthesized by pyrolyzing g-C3N4 embedded in carbon as highly efficient oxygen reduction electrocatalysts for primary Zn-air battery. Carbon 150, 475–484 (2019). https://doi.org/10.1016/j.carbon.2019.05.044

162. S. Wang, P. Xiong, J. Zhang, G. Wang, Recent progress on flexible lithium metal batteries: composite lithium metal anodes and solid-state electrolytes. Energy Storage Mater. 29, 310–331 (2020). https://doi.org/10.1016/j.ensm.2020.04.032

163. Y. Zhu, X. He, Y. Mo, Origin of outstanding stability in the lithium solid electrolyte materials: insights from thermodynamic analyses based on first-principles calculations. ACS Appl. Mater. Interfaces 7(42), 23685–23693 (2015). https://doi.org/10.1021/acsami.5b07517

164. Y. Zhu, X. He, Y. Mo. First principles study on electrochemical and chemical stability of solid electrolyte–electrode interfaces in all-solid-state Li-ion batteries. J. Mater. Chem. A 4(9), 3253–3266 (2016). https://doi.org/10.1039/C5TA08574H

165. Y. Huang, B. Chen, J. Duan, F. Yang, T. Wang et al., Graphitic carbon nitride (g-C3N4): an interface enabler for solid-state lithium metal batteries. Angew. Chem. Int. Ed. 59(9), 3699–3704 (2020). https://doi.org/10.1002/anie.201914417

166. Y. Lu, Z. Wen, J. Jin, Y. Cui, M. Wu, S. Sun, Mesoporous carbon nitride loaded with Pt nanoparticles as a bifunctional air electrode for rechargeable lithium-air battery. J. Solid State Electrochem. 16(5), 1863–1868 (2012). https://doi.org/10.1007/s10008-012-1640-8

167. Z. Sun, Y. Li, S. Zhang, L. Shi, H. Wu, H. Bu, S. Ding, G-C3N4 nanosheets enhanced solid polymer electrolytes with excellent electrochemical performance, mechanical properties, and thermal stability. J. Mater. Chem. A 7(18), 11069–11076 (2019). https://doi.org/10.1039/C9TA00634F

168. T. Liu, Y. Zhang, C. Chen, Z. Lin, S. Zhang, J. Lu, Sustainability-inspired cell design for a fully recyclable sodium ion battery. Nat. Commun. 10(1), 1965 (2019). https://doi.org/10.1038/s41467-019-09933-0

169. Z. Su, H.Y. Ling, M. Li, S. Qian, H. Chen, C. Lai, S. Zhang, Honeycomb-like carbon materials derived from coffee extract via a “salty” thermal treatment for high-performance Li-I2 batteries. Carbon Energy 2(2), 265–275 (2020). https://doi.org/10.1002/cen2.40

170. M. Sha, L. Liu, H. Zhao, Y. Lei, Anode materials for potassium-ion batteries: current status and prospects. Carbon Energy (2020). https://doi.org/10.1002/cen2.57

171. Y. Shi, G. Liu, R. Jin, H. Xu, Q. Wang, S. Gao, Carbon materials from melanin sponges for supercapacitors and lithium battery electrode materials: a review. Carbon Energy 1(2), 253–275 (2019). https://doi.org/10.1002/cen2.19

172. J. Chen, N.-G. Park, Materials and methods for interface engineering towards stable and efficient perovskite solar cells. ACS Energy Lett. 5, 180–185 (2020). https://doi.org/10.1021/acsenergylett.0c01240

173. J.-M. Fan, J.-J. Chen, Q. Zhang, B.-B. Chen, J. Zang, M.-S. Zheng, Q.-F. Dong. An amorphous carbon nitride composite derived from ZIF-8 as anode material for sodium-ion batteries. ChemSusChem. 8(11), 1856–1861 (2015). https://doi.org/10.1002/cssc.201500192

174. L. Chen, R. Yan, M. Oschatz, L. Jiang, M. Antonietti. K. Xiao, Ultrathin 2D graphitic carbon nitride on metal films: Underpotential sodium deposition in adlayers for sodium-ion batteries. Angew. Chem. Int. Ed. 59(23), 9067 (2020). https://doi.org/10.1002/anie.202000314

175. J. Liu, Y. Zhang, L. Zhang, F. Xie, A. Vasileff, S.-Z. Qiao, Graphitic carbon nitride (g-C3N4)-derived N-rich graphene with tuneable interlayer distance as a high-rate anode for sodium-ion batteries. Adv. Mater. 31(24), 1901261 (2019). https://doi.org/10.1002/adma.201901261

176. D. Adekoya, H. Chen, H.Y. Hoh, T. Gould, M.-S.I.T. Balogun, C. Lai, H. Zhao, S. Zhang, Hierarchical FeCo-NaN codoped hollow porous carbon with boosted electrochemical performances for sodium-ion batteries. Nano Lett. 18(8), 5137–5142 (2015)

177. X. Wu, S. Li, B. Wang, J. Liu, M. Yu, Free-standing 3D network-like cathode based on biomass-derived N-doped carbon composite as an advanced anode material for ultrastable potassium storage. ACS Nano 14(4), 5027–5035 (2020)

178. J. Liu, W. Li, L. Duan, X. Li, L. Ji, Z. Geng, K. Huang, L. Lu, L. Zhou, Z. Liu, A graphene-like oxygenated carbon nitride material for improved cycle-life lithium/sulfur batteries. Nano Lett. 15(8), 5137–5142 (2015)

179. X. Wu, S. Li, B. Wang, J. Liu, M. Yu, Free-standing 3D network-like cathode based on biomass-derived N-doped carbon/graphene/g-C3N4 hybrid ultrathin sheets as sulfur host for high-rate Li-S battery. Renew. Energy 158, 509–519 (2020).https://doi.org/10.1016/j.renene.2020.05.098

180. H. Zhang, Z. Zhao, Y.-N. Hou, Y. Tang, Y. Song, W. Xing, Z. Zhang, G. Cheng, Q. Jiu, Nanopore-confined g-C3N4 nanodots in N,S Codoped hollow porous carbon with boosted capacity for lithium-sulfur batteries. J. Mater. Chem. A 6(16), 7133–7141 (2018). https://doi.org/10.1039/C8TA0529J

181. X. Wang, G. Li, M. Li, R. Liu, H. Li, T. Li, M. Sun, Y. Deng, M. Feng, Z. Chen, Reinforced polysulfide barrier by g-C3N4/CNT composite towards superior lithium-sulfur batteries. J. Energy Chem. 53, 234–240 (2021). https://doi.org/10.1016/j.jechem.2020.05.036

182. Y. Gong, C. Fu, G. Zhang, H. Zhou, Y. Kuang, Three-dimensional porous C3N4 nanosheets@reduced graphene oxide network as sulfur hosts for high performance lithium-sulfur batteries. Electrochim. Acta 256, 1–9 (2017). https://doi.org/10.1016/j.electacta.2017.10.032

183. J. Wang, Z. Meng, W. Yang, X. Yan, R. Guo, W.-Q. Han, Facile synthesis of rGO/g-C3N4/CNT microspheres via ethanol-assisted spray drying method for high performance
lithium-sulfur batteries. ACS Appl. Mater. Interfaces 11(1), 819–827 (2018)

183. Y. Huangfu, T. Zheng, K. Zhang, X. She, H. Xu, Z. Fang, K. Xie, Facile fabrication of permselective g-3N4 separator for improved lithium-sulfur batteries. Electrochim. Acta 272, 60–67 (2018). https://doi.org/10.1016/j.electacta.2018.03.149

184. G. Angamuthu, D.B. Babu, K. Ramesha, V. Rangarajan, MoS2 anchored carbon nitride based mesoporous material as a polysulfide barrier for high capacity lithium-sulfur battery. J. Electroanal. Chem. 843, 37–46 (2019). https://doi.org/10.1016/j.jelechem.2019.05.006