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Graphitic carbon nitrides as electrode supporting materials for lithium-ion batteries: what lies ahead in view of the current challenges?

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Graphitic carbon nitride (g-C_3N_4) has emerged as a promising material for various applications, particularly in the field of energy storage systems. Among these systems, lithium-ion batteries (LIBs) have become the cornerstone of portable electronics and are increasingly being adopted for electric vehicles and renewable energy storage. However, the search for alternative electrode materials that can overcome the limitations of traditional graphite anodes and transition metal oxide cathodes remains a significant challenge. In recent years, g-C_3N_4 has attracted considerable attention due to its unique physicochemical properties, such as high electrochemical stability, tunable bandgap, large specific surface area, and excellent thermal and chemical stability. Also, the low cost, abundance, and environmental sustainability of g-C_3N_4 contribute to its suitability for next-generation LIBs. However, the successful utilization of g-C_3N_4 as an electrode material is hindered by several challenges. This paper aims to explore the challenges and future perspectives of utilizing g-C_3N_4 as a potential electrode material for LIBs, highlighting the potential benefits and drawbacks of integrating this material into the battery system.

KEYWORDS

graphitic, carbon nitride, lithium, rechargeable, electrode

1 Introduction

The Earth has an abundance of energy resources in the form of fossil fuels, most of which are being consumed at an exponential rate leading to rapid depletion of them. Despite, the depletion of fossil fuels, the energy requirement of the world will double by 2050 (Organization for Economic Co-operation and Development, 1999; International Energy Agency, 2050). Secondly, fossil fuels release greenhouse gases such as CO_2 and contribute to global warming. To mitigate these problems, scientists have been working on renewable and sustainable energy resources, such as solar energy, wind energy, tidal energy, etc., for a long time (Gong et al., 2015; Shamoon et al., 2022; Greening et al., 2023). Researchers developed many ways to utilize and store these energies in the form of electrical energy (Miller et al., 2015; Baig et al., 2022). The most powerful tool to store electrical energy is electrochemical devices such as batteries (Boruah et al., 2020), in which electrical energy can be stored in the chemical form and can be used directly from batteries when needed (Escudero-González & Amparo López-Jiménez, 2014; Fan et al., 2020). Thus, scientists have started to design and build high-performance rechargeable batteries (Yuan et al., 2022). Among the rechargeable batteries, LIBs have many advantages over other batteries. For example, LIBs are—1) more efficient and smaller in size as compared to other batteries, 2) very good in charge and discharge repeating cycle, 3) able to favor cordless charging, and 4) competent to prevent self-discharge (Liu et al., 2015; Zhou et al., 2018). LIBs possess good energy storage systems and have been extensively employed in many electric vehicles, transferable electronic gadgets, and electrical energy storage stations. Consequently, LIBs attracted huge marketable feat requiring more advancement such as high safety with high energy density capacity, low cost, and good reliability (Hu et al., 2017; Lu et al., 2019).

The main components of LIBs are a cathode (like—$LiCoO_2$), an anode (like—graphite), and an electrolyte (Encyclopedia of Sustainability Science and Technology, 2012). Cathode in LIBs behaves as a source of the Li-ions, once these ions pass through the electrolyte and accumulate in between the stacking layers of the anode, this phenomenon is called recharging of LIBs (Figure 1D). The LIBs follow the *vice versa* phenomenon of charging and discharging. In commercially available LIBs, graphite is widely acceptable as an anode as it has indispensable merits of high numbers of charging and discharging cycles, easy availability, good thermal and chemical stability, short voltage plateau, excellent chemical kinetics, and low cost (Safaei et al., 2018). In contrast, graphite anode has a low energy density rate (372 mAh g^{-1}) which does not equally align with the recently unveiled cathodes' energy density rate as these cathodes have a comparatively high rate (Fu et al., 2014). Besides, the propagation of lithium dendrite over the graphite anode is also a matter of acute safety concerns, such as catching fire (Lu et al., 2019). These shortcomings collectively restrict its application in high-performance LIBs. Subsequently, immense research has been done to mitigate these problems, and researchers designed and synthesized numerous materials to be used as an anode. However, the search for alternative electrode materials that can overcome the limitations of traditional graphite anodes and transition metal oxide cathodes remains a significant challenge (Liang et al., 2021). Among them, the two-dimensional graphite with nitrogen substitution at its maximum doping level provides a unique material called graphitic carbon nitride (g-C_3N_4) as a highly potential electrode material. Nitrogen doping of graphite provides a high coulombic cycle storage and reversible capacity. That is why, g-C_3N_4 has emerged as a promising material for various applications, particularly in the field of energy storage systems (Zhang H. et al., 2020; Wang et al., 2021; Tang et al., 2023). Also, this two-dimensional carbon-based material (g-C_3N_4) in comparison with graphite exhibits high oxygen-reducing reactivity, remarkable chemical stability, and high thermal stability. Furthermore, g-C_3N_4 has a tunable bandgap, high surface area, and excellent stability (Luo et al., 2019). These

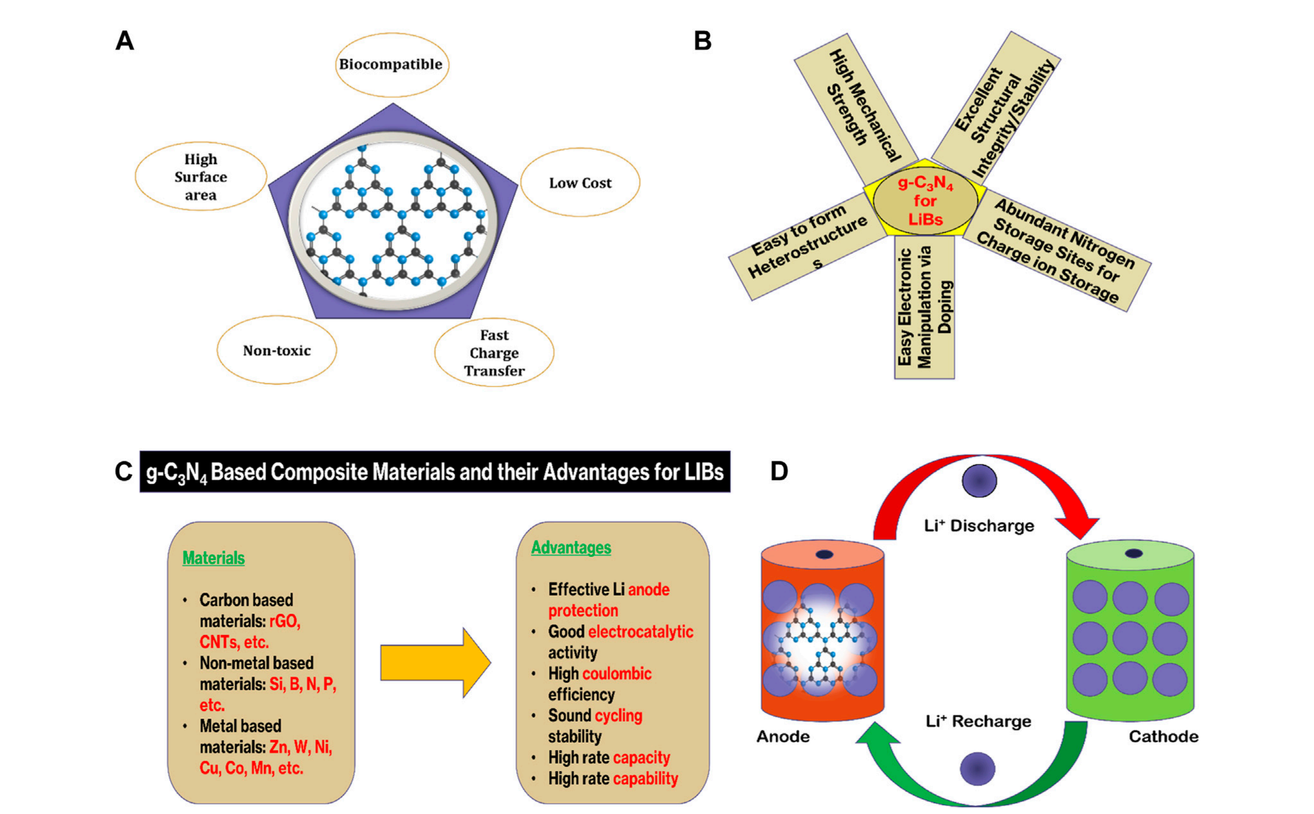

FIGURE 1
(A) g-C_3N_4 structure and its properties, **(B,C)** Advantages and physicochemical properties of g-C_3N_4 composites for LIBs, and **(D)** Charging and discharging of a g-C_3N_4-based LIB.

features all together make it an ideal candidate for batteries. In terms of metal-ion batteries (MIBs) like LIBs, g-C_3N_4 composites have shown superiority to some extent over other electrode materials (like: graphite, silicone, TiO_2) as demonstrated by Yadav et al., 2023, Maniyazagan et al., 2023, Nulu et al., 2022; Hankel et al., 2015, because its structure is based on the poly (triazine imide) webs. These webs contain pores to facilitate easy coordination with Li-ions (Zhao et al., 2015).

Instead of having of edge over other electrode materials, several challenges need to be addressed to realize the full potential of g-C_3N_4-based electrodes for LIBs. Among them, some of the major challenges compared to other electrodes are limited electrical conductivity, restricted cycling stability, and poor kinetics of LIBs (Gordon et al., 2023). Besides, large-scale production of g-C_3N_4 with cost-effective methods is still a quest. There is significant literature on the application of g-C_3N_4 in energy storage systems in the last decade, however, this manuscript is solely focused on the utilization of g-C_3N_4 as electrode material for LIBs. Also, this review article aims to explore those challenges and future perspectives of utilizing g-C_3N_4 as an electrode for LIBs, highlighting the potential benefits and drawbacks of integrating this material into the battery system. By addressing the key challenges and discussing the potential prospects, this analysis will shed light on the feasibility and relevance of g-C_3N_4-based LIBs in the quest for more efficient and sustainable energy storage solutions.

2 Physicochemical properties of g-C_3N_4

The physicochemical properties of g-C_3N_4 make it an attractive alternative to conventional electrode materials as shown in Figure 1. These physicochemical properties are chiefly influenced by the distinctive sheet structure of g-C_3N_4. The sheets of g-C_3N_4 are supposed to be made up of two primary unit rings—1) s-triazine (C_3N_4), and 2) tri-s-triazine (C_6N_7).

The s-triazine units are not as energetically favorable as the tri-s-triazine units (Cao et al., 2020). It was determined that the density functional theory (DFT) favors the thermodynamic stability of tri-s-triazine units over the s-triazine units (Kroke et al., 2002). Hence, it is broadly admitted that the tri-s-triazine units are the main constructing blocks of g-C_3N_4 sheets. Here, the key physicochemical properties of g-C_3N_4 that contribute to its suitability for LIBs have been conferred.

2.1 Thermal stability

The thermal stability of electrode materials is crucial to ensure reliable and safe operation. It is vital to develop such LIBs which can be operated at elevated temperatures for long-lastingEVs operation. For this purpose, g-C_3N_4 exhibits excellent thermal stability, with a decomposition temperature of around 600 °C (Liang et al., 2021). The high thermal stability of g-C_3N_4 ensures that it remains structurally intact and minimizes the risk of thermal runaway in LIBs (X. Liu et al., 2023). Thus g-C_3N_4 exhibits a paramount potential for thermally stable LIBs. However, there is a need of further research on commercialization of stable LIBs employing g-C_3N_4 as an electrode for elevated temperature environments as in EVs (Chen et al., 2019).

2.2 Electrochemical stability

g-C_3N_4 shows its structural integrity and performance over repeated charge-discharge cycles, exibhiting electrochemical stability. In, electrode materials undergo significant volume changes during cycling, which leads to mechanical degradation and capacity loss (Luo et al., 2019). However, plasma-induced highly nitrogen-deficient (ND) g-C_3N_4 electrode possesses a reasonably stable chemical structure resulting in retention of its performance even after 5,000 cycles (Sun et al., 2022). The remarkable chemical stability makes g-C_3N_4 a suitable candidate for long-lasting and high-capacity LIBs (Niu and Yang, 2018; Zhang J. et al., 2023).

2.3 Electrical conductivity

Pure g-C_3N_4 is a semiconductor with limited electrical conductivity. However, by incorporating dopants or adopting new synthesis methods, the conductivity of g-C_3N_4 can be significantly improved (Zhang X. et al., 2020). For instance, doping g-C_3N_4 with elements such as carbon or sulfur has been shown to enhance its conductivity, resulting in improved Li-ion battery performance (Hong et al., 2020). Likewise, recently developed technologies such as nitrogen-deficient g-C_3N_4 have a great potential for high-conductivity electrodes (Sun et al., 2022). The superior electrical conductivity will help the rapid and large-scale adoption of g-C_3N_4 in LIBs.

2.4 Specific surface area

The g-C_3N_4 exhibits a large surface area due to its unique porous structure (Huang et al., 2020), which consists of stacked layers with interlayer spacing that can accommodate a high density of lithium ions. The high surface area of g-C_3N_4 enhances the accessibility of lithium ions to the electrode material, facilitating faster charge-discharge processes and higher energy storage capacity (J. Zhang et al., 2015). The high surface areas also favor g-C_3N_4 as a highly potential electrode candidate for LIBs.

2.5 Sustainability and chemical stability

The carbon and nitrogen atoms in g-C_3N_4 form strong covalent bonds, which render the material resistant to chemical degradation. This stability ensures that the g-C_3N_4 electrode withstands solid-electrolyte-interphase leading to a prolonged lifespan of the electrode material, reducing the need for frequent replacements and lowering the environmental impact (Li et al., 2023). Additionally, g-C_3N_4 is composed of abundant elements, making it an attractive alternative to the limited and expensive resources used in traditional LIB materials, such as cobalt (Kong et al., 2018).

3 Application of g-C_3N_4 in LIBs

Especially to be used as an anode or cathode the composite structures of g-C_3N_4 can also be formed by combining with other materials (Rono et al., 2021), leveraging the advantages of both materials. Here are some specific applications of g-C_3N_4 in LIBs as an anode and cathode.

3.1 Auxiliary anode material

The pristine form of g-C_3N_4 has shown a higher theoretical capacity compared to graphite, with a capacity of around 524 mAh g^{-1} (Adekoya et al., 2021). The g-C_3N_4 unique electronic properties and porous structure allow for improved lithium-ion diffusion and enhanced cycling stability (Chen et al., 2017; Li et al., 2020). Therefore, as an anode, g-C_3N_4 composites of graphene (Wang et al., 2018), Nitrogen and Phosphorous (Tao et al., 2017), and salt of Se (Zhou et al., 2018), Zn (Joshi et al., 2018), Mn/Ni/Cu/Co (Zhang et al., 2019), Sn (Maniyazagan et al., 2023), WS_2 (Xu et al., 2022) exhibit improved specific capacities. While, 2,749 mAh g^{-1} is the highest initial discharge capacity so far reported for Sn/g-C_3N_4 composite anode (Le et al., 2022). So, it is deduced that g-C_3N_4 can be utilized as a composite with other materials to enhance its properties and performance.

3.2 Auxiliary cathode material

The g-C_3N_4 can also be employed as a cathode material in LIBs. It can serve as a host matrix for active materials, improving their electrochemical performance (Dutta et al., 2022). By incorporating active materials into the g-C_3N_4 matrix, the overall capacity and cycling stability of the cathode can be enhanced (Ramar and Wang, 2022). Similarly, the composites of g-C_3N_4 with graphene (Huang et al., 2016), porous carbon (Hong et al., 2020), nano sulfur copolymer (Yao et al., 2018; Tiwari et al., 2022), and salt of P (Zhang H. et al., 2020), and Cu (Li et al., 2023), show incredibly good cathodic properties. Hence, g-C_3N_4 also shows the characteristics of it being employed as a single electrode material or as support for other materials rendering them superior performance.

4 Challenges and future directions of g-C_3N_4 for LIBs

Despite the excellent merits of g-C3N4 (Ahmed and Maraz, 2023), as an electrode, it is facing many challenges to be used as a strong electrode material in LIBs, which are discussed in this section.

4.1 Challenges in utilizing g-C_3N_4 for LIBs

4.1.1 Limited electrical conductivity

One of the primary challenges associated with g-C_3N_4 as an electrode material for LIBs is its limited electrical conductivity (Ruby Raj et al., 2023). Compared to conventional carbon-based materials, such as graphite, g-C_3N_4 exhibits lower electron transport properties. This impedes the efficient movement of charge carriers during charge and discharge processes, leading to reduced battery performance (Luo et al., 2019). There have been efforts to improve the electrical conductivity of g-C_3N_4 by doping with other materials. However, enhancing the electrical conductivity of g-C_3N_4 without compromising its unique properties is still a critical challenge that needs to be addressed.

4.1.2 Poor kinetics of Li-ion intercalation

The inherently layered structure of graphite allows for easy intercalation of Li-ions, which enables the reversible charge storage mechanism in LIBs (Zhu et al., 2019). In the case of g-C_3N_4, however, the intercalation kinetics of Li-ions is slower (Li et al., 2022). This sluggish intercalation limits the battery's rate capability and affects charge-discharge performance (S. Wang et al., 2018). Developing strategies to improve the kinetics of li-ion intercalation within g-C_3N_4 is imperative for its successful integration into high-performance LIBs (Pathak et al., 2021).

4.1.3 Limited cycling stability

The g-C_3N_4-based electrodes demonstrate poor cycling stability, it is because they may experience mechanical stress, leading to structural degradation (Versaci et al., 2020). Li-ion insertion and extraction cause pulverization of the electrode. Further, slow kinetics of li-ion diffusion, unwanted side reactions between the electrode and the electrolyte leading to the formation of solid-electrolyte interface (SEI), and g-C_3N_4 may trap some lithium ions during cycling, reducing the reversible capacity of the electrode over time. These all phenomena contribute to the loss of cycling stability. Exploring novel approaches to mitigate structural deterioration and enhance the cycling stability of g-C_3N_4 electrodes is crucial for their industrial implementation as Sun et al., did, they built an anode material for LIBs by refilling of heteroatom in plasma-induced highly ND g-C_3N_4 (Sun et al., 2022).

4.1.4 Scalability and cost-effectiveness

For any promising material to find widespread application in LIBs, scalability and cost-effectiveness are key considerations (Xia et al., 2022; Liu et al., 2023). Currently, the synthesis of g-C_3N_4 lacks appropriate methods for large-scale production (Wang et al., 2019). Additionally, the cost of raw materials used for g-C_3N_4 synthesis is higher compared to conventional carbonaceous materials (Fang et al., 2016; Zou et al., 2016). Addressing these challenges is vital to ensure the viability and commercialization of g-C_3N_4-based LIBs.

4.2 Future directions

4.2.1 Designing hierarchical structures

The development of hierarchical structures can address the limited electrical conductivity of g-C_3N_4 (Xu et al., 2022). Introducing conductive additives or forming composite structures with conducting materials can improve the overall conductivity of the electrode. For example, incorporating carbon nanotubes or graphene into g-C_3N_4 matrices can enhance its electrical properties, leading to improved li-ion diffusion and charge transport within the electrode (Wang S. et al., 2023; Wang Y. et al., 2023).

4.2.2 Surface modification of g-C_3N_4

Surface modification strategies can be employed to enhance the kinetics of li-ion intercalation in g-C_3N_4-based electrodes. Functionalizing the g-C_3N_4 surface with active groups can facilitate li-ion diffusion and enhance the interaction between the electrode and electrolyte (J. Li et al., 2023). By careful selection and design of functional groups, improved intercalation kinetics and enhanced battery performance can be achieved (Choudhury et al., 2023).

4.2.3 Structural stability improvement

To improve the cycling stability of g-C_3N_4 electrodes, efforts must focus on stabilizing their structure during repeated lithiation and delithiation cycles (Li et al., 2017; Veith et al., 2013). Incorporating strengthening agents, such as carbon fibers or polymers, can provide mechanical support and alleviate structural distortion. Furthermore, exploring surface coatings and protective layers can prevent unwanted side reactions, reducing capacity fade, and improving long-term stability as reported by Maniyazagan et al.

4.2.4 Scalable synthesis methods

Developing scalable synthesis methods for g-C_3N_4 electrodes will facilitate their large-scale production. Innovative approaches like solvothermal, aerosol-assisted, or direct carbonization of nitrogen-containing precursors (Wang et al., 2020) can be explored (Villalobos et al., 2020). These techniques can promote efficient mass production of g-C_3N_4 electrodes, making them more commercially viable (Ramar and Wang, 2022).

4.2.5 Cost reduction

To enhance the cost-effectiveness of g-C_3N_4-based LIBs, efforts should be directed toward finding alternative and cheaper precursors for g-C_3N_4 synthesis (Han et al., 2019), which can be sustainably recycled or disposed off from used LIBs (Y. Liu et al., 2021). Moreover, innovative manufacturing techniques and optimization of fabrication processes can contribute to cost reduction for commercial production of LIBs with g-C_3N_4 (Zhang Y. et al., 2023; Degen and Krätzig, 2023).

5 Conclusion

In conclusion, the g-C_3N_4 exhibits various physicochemical properties that make it a promising material for LIBs. Its high thermal stability, large specific surface area, chemical stability, and environmental sustainability contribute to its suitability for LIBs, as discovered recently. These exceptional properties of g-C_3N_4 make it a favorable material for diverse functions in LIBs, including anodes, cathodes, composites, electrolyte additives, and separator coatings. Also, its incorporation in LIBs can lead to improved energy storage performance, enhanced cycling stability, and increased safety. Despite the challenges faced by g-C_3N_4 for LIBs, concerted efforts in addressing the limited electrical conductivity, poor intercalation kinetics, scalability, and cost-effectiveness can pave the way for its successful integration. Additionally, continued research and development in synthesizing and optimizing g-C_3N_4-based electrodes for high-performance and environment-friendly LIBs can contribute to a more sustainable and efficient energy storage future. g-C_3N_4 can emerge as a competitive material for next-generation LIBs, by designing hierarchical structures, surface modification, improving structural stability, developing scalable synthesis methods, and reducing production costs.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary material, further inquiries can be directed to the corresponding authors.

Author contributions

MS: Investigation, Writing–original draft, Writing–review and editing, Data curation, Validation. ZC: Investigation, Validation, Writing–review and editing. BH: Investigation, Writing–review and editing, Project administration, Supervision. TY: Investigation, Project administration, Supervision, Writing–review and editing. JC: Investigation, Project administration, Supervision, Writing–review and editing, Conceptualization, Funding acquisition, Resources, Visualization, Writing–original draft.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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