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**Review Article** 

## A Comprehensive Review of the Multidimensional Nanostructural Properties of Graphene Carbon Nitride as a Supercapacitor Electrode

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#### ABSTRACT

This abstract offers a comprehensive review of recent advancements in Graphene Carbon Nitride (GCN) as a highly promising electrode material for supercapacitors. GCN boasts exceptional advantages, including abundant availability, a metal-free composition, high nitrogen content, and remarkable environmental sensitivity. These unique characteristics have positioned GCN at the forefront of research in energy storage and supercapacitor electrode materials. However, despite its potential, GCN faces challenges concerning limited specific capacity and energy density. To address these limitations, this review, as the first and most comprehensive in its field, focuses on innovative and novel development methods, particularly the strategic formation of nanostructures in 1, 2, and 3 dimensions. A notable finding of this review is the tremendous promise of 3D structures in enhancing the electrochemical properties of GCN as a supercapacitor electrode. A critical research gap in other review articles is the absence of comprehensive and innovative literature investigating nanostructures (1D, 2D, and 3D) with novel synthesis methods for using GCN as a supercapacitor electrode. This underscores the pressing need for further scholarly investigation in this area, as addressed by this review article. Overall, this professional review not only provides a comprehensive overview of advancements in GCN as a supercapacitor electrode material but also offers valuable guidance for researchers in the field. It highlights the importance of utilizing environmentally friendly synthesis techniques for fabricating multidimensional nanostructures, illuminating novel research directions and pioneering investigations. This empowers researchers to advance the utilization of GCN in energy storage applications.

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### 1. INTRODUCTION

The increasing global demand for energy and the environmental consequences of fossil fuel depletion have intensified the search for clean, sustainable, and economically viable energy solutions (Mustaqeem, Naikoo, Rahimi, et al., 2022; Mustaqeem, Naikoo, Yarmohammadi, et al., 2022; Naikoo et al., 2023). Solar, wind, and tidal power represent the foremost renewable energy sources due to their abundant

availability and significant potential. The widespread adoption and advancement of clean energy and development programs have been hindered by a persistent challenge: the intermittent availability of renewable energy sources. An illustrative example is solar power, traditionally limited to daylight hours. However, this limitation can be substantially addressed through energy storage solutions, elevating the significance of energy storage as a globally paramount concern.

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The quest for efficient and reliable energy storage technologies has emerged as a compelling and pressing issue in the pursuit of a sustainable and resilient energy future. Furthermore, deliberate efforts have been made to construct contemporary energy storage technologies that cater to the requirements of current energy consumption and the anticipated sustainable energy infrastructure of the future. In this regard, electrochemical energy storage devices, which efficiently capture and store renewable energy sources, assume a crucial role. These devices are essential components in addressing the imperative need for energy storage, facilitating the seamless integration of intermittent renewable energy generation into the grid, and laying the foundation for a resilient and sustainable energy ecosystem.

Supercapacitors, also known as ultracapacitors, have garnered significant research attention due to their distinctive attributes, including rapid charge-discharge behavior, high power densities, and remarkable cyclic stability. These energy storage devices offer a compelling solution with the potential to revolutionize the field of energy storage. The exceptional performance characteristics of Supercapacitors make them a captivating avenue for addressing the pressing demands of modern energy systems, enhancing power delivery capabilities, and enabling novel applications in various industries. Supercapacitors have a structure composed of two electrodes, an electrolyte, and current separation and collection components (Antil et al., 2022; Farrag et al., 2018; Khan et al., 2020; King et al., 2017; Meroueh et al., 2020). The schematic structure of Supercapacitors can be seen in Figure 1.

Supercapacitors hold promise for advanced energy storage solutions with their rapid charge-discharge behavior, high



power densities, and prolonged cyclic stability. The electrode is responsible for storing the charge in Supercapacitors and is the main and most important component of the Supercapacitor structure that has been the most researched. According to their method of charge storage, the electrode materials used in Supercapacitors realization are often divided into three categories: (I) electric double-layer capacitors (EDLCs), (II)

pseudocapacitive Supercapacitors (III) batteries-like Supercapacitors. **EDLCs** (Electric In Double-Laver Capacitors), the interface between the electrode and electrolyte is where charges are physically adsorbed through non-Faradaic mechanisms. Active substances such as Reduced Graphene Oxide (RGO), single and multi-walled carbon nanotubes, and activated carbon derived from biological sources are commonly employed in this context. These materials, often utilized as anode electrode materials in Supercapacitors, offer reliable power sources with exceptional properties (Isacfranklin et al., 2020; Isacfranklin et al., 2021; Karuppaiah et al., 2020). Graphene carbon nitride is recognized as one of the most groundbreaking materials in the category of carbon-based materials, attracting significant attention due to its intrinsic properties for utilization as a supercapacitor electrode. The presence of nitrogen within the structure of graphene carbon nitride is regarded as one of its paramount intrinsic characteristics, endowing it with superior utility compared to other carbon-based materials. This nitrogen incorporation plays a pivotal role in enhancing the electrochemical properties of graphene carbon nitride. The GCN structure part elaborates on this aspect.

The Faradaic redox method is employed to store charges within battery-type electrode materials, maintaining a constant potential window at the interface between the bulk electrode and electrolyte. Key constituents, including CO<sub>3</sub>O<sub>4</sub>, NiO, Mn<sub>2</sub>O<sub>3</sub>, and Ni-Co-layer hydroxide (Chen et al., 2014; Sriram et al 2021), offer reliable energy storage capabilities. Conversely, pseudocapacitive electrode materials demonstrate a continuously changing potential window, facilitating charge accumulation through faradaic reactions at the material's surface. Chalcogenide-based materials like FeS, Ni<sub>3</sub>S<sub>4</sub>, Cu<sub>2</sub>S, MoS<sub>2</sub>, and Bi<sub>2</sub>S<sub>3</sub> have gained significant attention as cathode and anode materials for energy storage and conversion applications (Choi et al., 2020). High-performance electrodes possess key attributes essential for optimal performance. Firstly, they integrate active materials with significantly enhanced specific surface areas, allowing for a greater influx of electrolyte ions and promoting efficient redox reactions. Secondly, a meticulously designed mesopore structure is vital in facilitating the seamless transfer of electrolytes. Lastly, electrodes characterized by high electrical conductivity and an expansive potential window contribute to exceptional specific capacitance, remarkable rate capability, and outstanding cycle stability, making them highly appealing for applications involving repeated charge and discharge cycles. Carbon-based materials emerged as the trailblazers in the realm of conductive materials for supercapacitor electrodes. Their exceptional electrochemical properties, including a high specific surface area, lightweight composition, unparalleled stability, costeffectiveness, non-toxicity, and abundant manufacturing sources, have placed them at the forefront of supercapacitor applications. Carbon nanotubes, graphene, fullerenes, and other carbon compounds can be ingeniously fashioned into multidimensional and multifunctional structures, elevating their electrical and mechanical prowess (Ali et al., 2021; Ghanem et al., 2021; Liang et al., 2015; Lu et al., 2017). GCN, an enticing 2D semiconducting carbon material free from metals, boasts a graphite-like structure, remarkable thermal stability, high nitrogen concentration, chemical resilience, and exceptional mechanical strength. This captivating material merges intriguing electronic properties with superb material characteristics. GCN emerges as a compelling candidate with

immense potential across diverse applications (Idris & Devaraj, 2019). Due to the nitrogen heteroatom's ability to enhance capacitance, enable mixed charge storage methods, and increase surface reactivity compared to undoped carbon counterparts, GCN has been extensively studied as a promising carbon-based compound for energy storage purposes. Structurally, GCN-based supercapacitor materials offer notable advantages. Firstly, GCN possesses a theoretical nitrogen content of 57.1%, facilitating electron aggregation and pseudocapacitance within the electrode materials. Secondly, a significant portion of N atoms in GCN exists as pyridine nitrogen, potentially enhancing electrode pseudo-capacitance. Furthermore, positively charged quaternary nitrogen aids in electron transport, while graphite nitrogen bound to three carbon atoms enhances material conductivity (Lin et al., 2015; Luo et al., 2019; Yu et al., 2020). Lastly, GCN can be regarded as a nitrogen-rich carbonaceous material with robust cycle stability for supercapacitor utilization. Despite the ideal characteristics attributed to GCN, this captivating material faces inherent limitations that impede its further development and utilization as a supercapacitor electrode. The limited conductivity and low specific surface area of GCN stand as two crucial factors that hinder its potential as a supercapacitor electrode.

This comprehensive review article extensively explores cutting-edge structural morphologies. The primary aim is to unlock the full potential of GCN as an electrifying electrode material for supercapacitors, pushing its intrinsic characteristics to unprecedented heights. What sets this review apart is its meticulous analysis and comprehensive examination of the benefits and drawbacks of various nanostructures for GCN as an electrode material in supercapacitors. This distinctive approach has assisted researchers in identifying the ideal development method that seamlessly aligns with their research goals.

Through a thorough investigation into the advantages and limitations of diverse development methods for GCN, researchers gain invaluable insights, empowering them to make astute decisions tailored to their specific objectives. For instance, the exploration of environmentally sustainable synthesis techniques for the multi-dimensional structural synthesis of GCN as a supercapacitor electrode represents an underexplored area of research and a notable research gap. This article provides relevant solutions to broaden scholarly contributions to this field.

According to Figure 2, the structure of GCN is initially discussed in this review article. The "GCN Structure" section delves into the structural and intrinsic features of GCN. In the "Morphology Control" section, the unique features of each 1, 2, and 3-dimensional structure in GCN are discussed, along with the benefits and drawbacks of the techniques used to create these nanostructures.



Figure 2. Main items discussed in this review article

In conclusion, the article presents multiple proposals to determine the appropriate synthesis method and development mechanism. It is worth noting that collecting and analyzing various development methods for GCN as a supercapacitor electrode with such comprehensive details and expressing these findings in a simple and transparent manner distinguishes this review article from others in the field.

As depicted in Figure 2, this review article undertakes a comprehensive examination of the GCN structure. The discussion then extends to cover various development strategies. The section on morphology control explores the distinctive attributes conferred by 1, 2, and 3-dimensional structures in GCN, while also scrutinizing the benefits and drawbacks associated with the techniques employed for nanostructure fabrication. Finally, the concluding section presents multiple proposals to determine the most suitable development mechanism.

One of the major challenges in crafting this article was amalgamating scattered research and establishing a cohesive connection. This involved ensuring a smooth and uniform expression between the characteristics of the multidimensional states of the graphene carbon nitride structure and the electrochemical parameters of this material. It's crucial to emphasize that this review article stands out due to its meticulous collection and comprehensive analysis of various development methods for GCN as a supercapacitor electrode. The information is presented clearly and transparently, making this article an essential resource that distinguishes itself from others in the field.

#### 2. GCN structure

To enhance the electrochemical efficiency of GCN as a supercapacitor electrode material, acquiring detailed information about its structural characteristics is crucial. This review article commences by introducing the intricate and complex structural features of GCN. Notably, Berzelius conducted a seminal study on GCN, investigating covalent carbon nitride using heptazine units as the primary structural constituents. Additionally, first-principle calculations have provided fascinating insights, revealing the existence of seven distinct phases of GCN. One particular phase exhibits a remarkable energy value of 4.13 eV. These captivating findings form the cornerstone of this article, offering a professional and engaging exploration of the structural aspects of GCN. The diverse phases of GCN, including  $\beta$ -CN, pseudo cubic GCN, gtriazine, gh-heptazine, g-triazine, α-CN, and cubic CN, showcase intriguing properties (Sevilla & Mokaya, 2014; <u>Wang et al., 2012</u>). The renowned  $\beta$ -GCN crystalline phase is of particular interest due to its diamond-like hardness and low compressibility. This unique combination holds promise for applications in durable materials and deformation-resistant systems, presenting an exciting avenue for scientific inquiry and potential technological breakthroughs. β-GCN, resembling graphite in structure, features a nitrogen lattice with partial substitution, setting it apart from the typical carbon lattice. This unique arrangement holds scientific intrigue, offering insights into advanced materials and their potential applications. By delving into the intricacies of this nitrogen-substituted lattice, researchers can uncover fundamental principles governing material behavior and drive innovations in energy storage and catalysis. Exploring the structure of  $\beta$ -GCN paves the way for exciting scientific discoveries and the development of customized materials. GCN, with its intriguing combination of two and three-dimensional structures, exhibits characteristics of an n-type semiconductor, owing to its high concentration of nitrogen. This concentration is primarily attributed to the presence of nitrogen pyridinic and graphitic elements, contributing to the material's fascinating electronic properties. Significantly, GCN stands out as the most stable form among carbon nitrides (Thomas et al., 2008). In principle, the amalgamation of nitrogen and carbon has resulted in concurrent enhancement of electrochemical properties and structural stability, with nitrogen playing a pivotal role in improving electrochemical properties, while carbon exhibits excellent structural stability owing to its intrinsic characteristics. This stability arises from the interlayer bonding between adjacent layers, mediated by van der Waals forces measuring 0.036 eV A<sup>-2</sup>. Structurally reminiscent of graphite, the bulk GCN consists of multiple layers held together by these cohesive forces (Zhang et al., 2016).

These intriguing similarities between GCN and graphite open up exciting avenues for exploring its unique properties and potential applications in the realm of advanced materials and electronic devices. Interestingly, during the calculations, a fascinating revelation emerged regarding the inter-planar distance. It was found that tectonic units such as tri-s-triazine (heptazine,  $C_6N_7$ ) and s-triazine (CN) could potentially serve as allotropes of GCN. These compounds are believed to be intricately interconnected by trigonal nitrogen atoms, forming substantial co-planar networks within the p-conjugated planar layers (see Figure 3)(Tahir et al., 2013).

This discovery offers a captivating glimpse into the structural possibilities of GCN, showcasing its potential for diverse forms and intricate arrangements. By unraveling the intricate tectonic units and their interconnections, scientists can further explore the unique properties and applications of these GCN allotropes, paving the way for exciting advancements in materials science and beyond. In recent times, there has been a surge of interest in density functional theory (DFT) calculations as a potent and essential tool for examining the geometric structure of GCN. One notable study by Zhu et al. provided a summary of specific DFT research developments, encompassing both s-triazine and tri-s-triazine units(Zhu et al., 2018). Another research conducted by Kroke et al. revealed that a tri-s-triazine-based GCN structure exhibited 30 kl mol<sup>-1</sup> higher stability compared to an s-triazine-based structure(Zhu et al., 2019). These findings shed light on the molecular architecture and stability of GCN, offering promising avenues for future advancements in its design and application. The energetically advantageous tri-s-triazine rings emerge as the preferred building blocks for GCN. However, during thermal polymerization, incomplete polymerization of residual cyanamide or amino functional groups often occurs, resulting in the presence of unavoidable faulty components. These components, characterized by one-dimensional melon chains, pose a significant obstacle to the ideal development of the GCN structure. The polymerized form of GCN primarily consists of carbon, nitrogen, and residual hydrogen derived from unconstrained amino functionalities. Remarkably, the average C/N atomic ratio typically measures 0.72, slightly lower than the predicted value of 0.75 (Rahman et al., 2021; Thomas et al., 2008). This disparity underscores the intricate nature of GCN's polymerization process, necessitating further exploration and refinement to achieve the desired structural integrity. For the purpose of elucidating the impact of the intrinsic properties of graphene carbon nitride on electrochemical parameters,

reference can be made to the following two studies. This approach aims to enhance transparency in the examination of these effects. In the first study, Mahrokh Nazari et al synthesized pure aerogel GCN, resulting in an electrode that demonstrated a specific capacity of  $240 \text{ Fg}^{-1}$ . (Nazari et al., 2021) Conversely, in another study conducted by Qinghong Wang et al, an Fe<sub>3</sub>O<sub>4</sub>/graphene hybrid composite, which involved a more intricate and protracted synthesis process, exhibited a specific capacity of  $220 \text{ Fg}^{-1}$ . (Wang et al., 2014) These two studies represent a fraction of the broader research corpus that validates the impact of graphene carbon nitride's core properties on electrochemical parameters.



Figure 3. Schematic of different structures of GCN

#### 3. Enhancing the Intrinsic Properties of GCN

GCN: Unveiling Innovations. GCN, despite possessing ideal inherent properties, exhibits certain limitations that have constrained its development programs. These limitations encompass inadequate conductivity, low specific surface area, and other notable shortcomings. To overcome these challenges, substantial endeavors have been dedicated to resolving these issues. For instance, Chen and colleagues have successfully created a potentially reversible GCN anode with an impressive lithium storage capacity of 2753 Fg<sup>-1</sup> after 3000 cycles(Chen et al., 2017). In another study, Liqi Bai et al. reported on the functional capabilities of transition metal oxides in supercapacitors and the promising potential of GCN heterojunctions in photocatalysis(Bai et al., 2022). In general, the control of architectures has shown significant improvement in the electrochemical storage performance of GCN-based materials in batteries and supercapacitors. Recent studies, as depicted in Figure. 4 (Dong et al., 2017; Li et al., 2016), have yielded a plethora of GCN-based materials, particularly through the utilization of novel synthesis techniques. These advancements hold immense promise in the field of electrochemical energy storage, opening avenues for enhanced capabilities and diverse applications.



# Figure 4. Advantages of developed GCN against the limitations of the pure GCN state

However, there have been only a few reviews that have specifically addressed the area depicted in Figure 5. What distinguishes this review from other articles in the field is its comprehensive examination of the strategies mentioned in Figure 5. By thoroughly analyzing these strategies, this review aims to provide valuable insights and contribute to the existing knowledge base in this research area, following the conventions and rigor expected of a comprehensive review paper. Both graphite and GCN exhibit three-dimensional structures. Typically, nitrogen-rich precursors such as melamine, dicyandiamide, thiourea, or urea are commonly pyrolyzed to produce bulk GCN (Mo et al., 2018; Rono et al., 2021). However, GCN faces challenges, including limited quantum efficiency, a small pore size  $(0.1 - 0.5 \text{ cm}^3\text{g}^{-1})$ , and a modest surface area. These limitations necessitate innovative approaches to enhance the performance of GCN-based materials. Due to the mechanisms of charging and discharging electrical energy in supercapacitors, both electric double-layer capacitors (EDLCs) and pseudo-capacitors, having a high surface area is considered a crucial parameter for achieving a higher specific capacity. To address these limitations, researchers have discovered methods to transform bulk GCN into various dimensional forms, such as 1D, 2D, and 3D structures, nanosheets, nanotubes, core shells, and more (Gao et al., 2019). These transformations aim to provide additional active sites and increase the exposed surface area.



Figure 5. Summary of issues raised in the GCN with nanostructure

#### 3.1. GCN with nanostructure

The synthesis of materials with different nanostructured modes, aimed at improving their performance, is currently one of the most widely debated subjects worldwide. Researchers from

across the globe are actively involved in exploring and discussing various strategies in this field. GCN can be synthesized with various nanostructures, including 0D, 1D, 2D, 3D, nanosheets, nanorods, nanotubes, nanoneedles, and more, demonstrating exceptional performance as electrode material in supercapacitors. Extensive research has been conducted on this subject, encompassing a wide range of studies and scientific investigations. For instance, Faheem K. Butt employed an innovative microwave-assisted method to fabricate twodimensional GCN (Butt et al., 2020). The resulting nanostructures of GCN exhibited a specific capacitance of 287 Fg<sup>-1</sup>, showcasing their exceptional electrochemical performance. Additionally, the electrochemical behavior of pure GCN was assessed using a quartz crystal nano balance, revealing specific capacitance of 10  $Fg^{-1}$ . The synthesis of GCN with such nanostructures has yielded improved parameters, including specific surface area, conductivity, and active sites. These advancements underscore the potential of tailored GCN structures in optimizing the performance of supercapacitors, making them an attractive avenue for further exploration and development. Enhancing the specific surface area on 1D, 2D, and 3D structures has amplified the interfacial area between the electrode and the electrolyte, thereby augmenting the energy storage within the electrode. Consequently, it has elevated the electrical conductivity, expedited the kinetics of electrochemical reactions, and ultimately accelerated the charging process.

However, the currently available synthetic methods for obtaining GCN with specific morphologies have certain limitations, such as lengthy processing times, high costs, and the use of toxic solvents. In this section, a comprehensive analysis is provided, elucidating the benefits and drawbacks of employing 1D, 2D, and 3D structures of GCN as electrodes for supercapacitors. Additionally, novel methods utilizing nontoxic solvents for GCN synthesis are discussed, highlighting their potential to overcome the aforementioned limitations. Furthermore, the efficiency of these alternative approaches is examined in comparison to various other synthesis techniques. This section offers valuable insights into optimizing the morphologies of GCN for improved supercapacitor electrode performance while considering environmental and cost-related factors associated with the synthesis process.

#### 3.1.1. 0D structures

The 0-dimensional structure has been recognized as an attractive architectural design capable of endowing different materials with ideal properties. Notably, GCN, when engineered in a zero-dimensional (0D) configuration, has garnered increasing interest due to its desirable qualities, including chemical stability, a significant specific surface area, biocompatibility, and a modest band gap. These advantages have sparked a growing fascination with 0D GCN and its potential applications. Moreover, the exceptional attributes of the 0-D GCN make it an appealing choice as an electrode material for supercapacitors (Ngo et al., 2019). Ping Zhao, Bo Jin, et al.., accomplished a noteworthy feat in their research by fabricating recyclable hybrid aerogels combining RGO/GCN quantum dots (QDs) )(Zhao et al., 2021). This innovative approach resulted in aerogels that exhibited improved photocatalytic function. Moreover, owing to their mesoporous structure, the RGO/GCN QDs possessed a substantial surface area and displayed solid catalytic activity. Notably, the exceptional advantages of these materials, including their excellent specific surface area and chemical stability, have proven pivotal in advancing the development programs of supercapacitors (Niu et al., 2021; Wang et al., 2016). Additionally, the utilization of the 0D structure of GCN, either in its pure form or as a composite, presents an intriguing option for electrode fabrication that has received comparatively less attention. The 0D GCN, owing to its exceptional structural properties, holds the potential to enhance the functional parameters of supercapacitors. Consequently, this avenue of research emerges as an attractive and promising area worthy of further exploration.

#### 3.1.2. 1D structures

The investigation of 1D structures has been a primary focus for researchers in the realm of electrode manufacturing, given their numerous advantages. A wide range of materials, including carbon-based materials, have been successfully synthesized in this structural form. Notably, carbon-based 1D materials, such as graphene-based fibers, carbon fibers, and carbon nanotubes, offer remarkable qualities that contribute to enhanced capacitance and electrical conductivity. These materials possess attributes such as a large surface area, low mass density, and high tensile strength, making them highly desirable for the development of advanced materials with superior performance.

All of these parameters have had a significant impact on the performance of supercapacitors, leading to extensive research on 1D carbon-based materials. For example, Renjie Zhang successfully fabricated 1D nano needle-shaped carbon nano bars (CNB) (Zhang et al., 2022). Zhang's research revealed a specific region with a considerable specific surface area of 879.18 m<sup>2</sup>g<sup>-1</sup> for the CNB material. Remarkably, when utilized as a flexible electrode, the CNB exhibited outstanding areal capacitance of 2245Fcm<sup>-2</sup> at 1 mAcm<sup>-2</sup>. Additionally, the flexible asymmetric device demonstrated exceptional stability over 20,000 cycles (with 99.5% retention) and exhibited a superior Power density of 0.52  $\text{mWem}^{-2}$ . These findings highlight the immense potential of 1D carbon-based materials in enhancing the performance of Supercapacitors, emphasizing the significance of continued research in this field. GCN, being a carbon-based material, demonstrates ideal properties when employed in 1D structures. For instance, 1D GCN-based nanotubes exhibit a higher specific surface area and provide a greater number of active catalytic sites (Bakr et al., 2019; Kumar et al., 2017; Mahzoon et al., 2018). Moreover, these nanotubes possess both an elongated axial length and a reduced radial length, thereby reducing the distance for carrier migration. The 1D structure's higher specific surface area and shorter radial length, in comparison to 0D structures, make it advantageous for efficient ion transfer. As a result, this structure holds significant potential for applications in capacitors and supercapacitors, leading to extensive research in this area. In a noteworthy research endeavor by S.V. Prabhakar Vattikuti et al., a ternary electrode substance for supercapacitors was successfully fabricated, combining Carbon/CuO nanospheres with GCN nanosheets (Vattikuti et al., 2018). This ground-breaking study yielded exceptional results, as the composite demonstrated a specific capacitance of 247  $Fg^{-1}$  with a current density of 1  $Ag^{-1}$  in 0.5 M NaOH. Impressively, even after undergoing 6000 cycles, the composite retained over 92.1% of its initial capacitance. These remarkable findings underscore the tremendous potential of composite materials in enhancing both the performance and longevity of

Supercapacitors, offering exciting prospects for further investigation.

Materials based on 1D GCN can be produced utilizing both the hard template and soft template techniques. However, it is widely believed that creating 1D GCN-based materials through melamine self-assembly represents a superior method. This approach offers several advantages, including reduced solvent usage and higher stability of the final product. By employing melamine self-assembly, researchers can achieve the desired 1D structure while minimizing solvent consumption and ensuring the enhanced durability of the resulting materials. In another research study, a comprehensive synthesis process for CO<sub>3</sub>O<sub>4</sub>/Ag/GCN in a 1D form is summarized as follows: Initially, spiral AgNO3-melamine nanowires were fabricated by combining melamine and AgNO<sub>3</sub> (Chen et al., 2019). Subsequently, these nanowires were collaboratively assembled in situ by incorporating melamine and cyanuric acid. Next, an amide nitrogen complexing agent, along with CO(NO<sub>3</sub>)<sub>3</sub>, was introduced to establish a linkage between  $CO_3$  and melamine/cyanuric acid. Finally, during a nitrogen-containing calcination at 550 °C, melamine and cyanuric acid polymerized along an Ag<sup>+</sup> melamine nanowire, resulting in the formation of a rod-like structure of GCN. This intricate procedure demonstrates the meticulous approach employed to successfully produce the desired CO<sub>3</sub>O<sub>4</sub>/Ag/GCN material in a one-dimensional form. The controlled synthesis techniques employed in this process highlight the significance of precise methodology in achieving unique and well-defined structures. Exfoliation stands as an innovative technique for creating GCN with a nanostructure. This strategy encompasses various methods, including ultrasonic exfoliation, thermal exfoliation, acid-assisted in-situ exfoliation, gas-bubbling exfoliation, and gas exfoliation, among others. What distinguishes this synthesis method from others is its higher yield and straightforward operational procedure. Furthermore, numerous research studies have been conducted to investigate and advance this method, highlighting its importance and potential for further exploration. For example, Chao Li successfully fabricated NiFeO4/GCN with a core-shell hollow structure, where GCN was synthesized using the thermal exfoliation method. The specific capacities of prNFO/GCN were measured at 85.27 mAhg<sup>-1</sup>, and a specific capacity of 57 mAhg<sup>-1</sup> could be maintained at 20 Ag<sup>-1</sup>, resulting in an impressive 67.2% retention for prNFO/GCN. These findings highlight the significant potential of prNFO/GCN as an electrode material, exhibiting enhanced specific capacity and excellent performance at high current densities. Chao Li's research contributes valuable insights into the development of advanced materials for energy storage applications(C. Li et al., 2021). In other study, Arun Kumar et al. synthesized free acid-etched graphitic carbon nitride (TGCN) nanosheets with optimized PANI nanofibers and this composite shows specific capacitance of 298.31 Fg<sup>-1</sup> (Kumar & Khanuja, 2021). Obtaining such a special capacity for a two-component composite that only includes polymer and carbon materials is considered a great achievement and it has been promising that such new synthesis methods can lead to very ideal results.

The exceptional electrochemical performance observed in this study can be ascribed to two key factors: partial reduction and incorporation of core-shell hollow spherical constructions, both of which were optimized to enhance overall performance. The investigations carried out in this article have revealed that the increased porosity and specific surface area of 1D GCN, along with their mutual influence on conduction and electron transfer, have significantly improved the electrochemical parameters when compared to 0D materials. These findings emphasize the critical role of optimizing the structural characteristics of GCN in achieving remarkable electrochemical performance, thus opening up promising avenues for further advancements in this field.

#### 3.1.3. 2D structures

2D structures offer a precise and promising avenue for the utilization of carbon-based materials. Specifically, 2D GCN nanosheets, consisting of mono or few-layered GCN, bear a striking resemblance to graphene in terms of their structure. However, unlike graphene, these nanosheets are characterized by the connection of Tris-triazine units through amino groups, which results in the presence of periodic lattice vacancies and dangling hydrogen within the C and N layers (Barman & Sadhukhan, 2012; She et al., 2014; Wang et al., 2021). This distinctive arrangement imparts added complexity and versatility to the properties of 2D GCN nanosheets, thereby highlighting their significant potential for diverse applications in the realm of advanced materials. GCN nanotubes with high aspect ratios present an intriguing 2D structure within the realm of GCN, offering unique advantages in electron migration facilitation, increased active sites and surface area, enhanced electrical conductivity, and improved redox reactions between the electrolyte and electrode, as compared to bulk GCN (Zou et al., 2020). This review delves into the most recent research on this captivating nanotube structure, providing valuable insights into its potential applications and highlighting its promising attributes across various fields.

Wang et al. utilized calcium carbonate as a rigid template to create mesoporous GCN, which was subsequently removed using hydrochloric acid (Wang et al., 2015). The resulting mesoporous GCN nanotubes exhibited an impressive BET surface area of 38.6 m<sup>2</sup>g<sup>-1</sup>, highlighting the significant impact of structural modification on enhancing the specific surface area of GCN. In another study, Mingjie Li demonstrated that the exceptional electrochemical performance of materials based on cobalt diselenide was attributed to the synergistic interaction between GCN and carbon nanotubes (M. Li et al., 2021). This effectively improved synergistic collaboration their electrochemical properties. These findings emphasize the crucial role of structural engineering and the exploration of synergistic effects in optimizing the performance of advanced materials. They provide valuable insights for future research and development in the field. The wrinkled surface observed in self-assembling COSe<sub>2</sub> during slow thermal exfoliation is attributed to the abrupt edge bend, which results in a porous skeleton and a substantial inner surface area. This unique design not only enhances the activity levels for redox reactions but also provides a broader contact point for the adsorption and desorption of electrolyte ions. Moreover, the presence of gullies between the wrinkled GCN nanosheets helps alleviate strain during repeated charge cycles, reducing structural damage to the electrode and promoting desired cyclic stability. Impressively, the COSe2/CNTs/GCN composite exhibits a superior specific capacitance of 445.4 mAhg<sup>-1</sup> at 1 Ag<sup>-1</sup>, while the assembled asymmetric supercapacitors based on COSe<sub>2</sub>/CNTs/GCN deliver a high energy density of 7.1 whkg<sup>-1</sup> These findings underscore the remarkable potential of this composite material, offering both exceptional performance and cyclic stability, thus paving the way for advanced energy

storage applications. K.C. Devarayapalli et al. reported on a remarkable example of composite structures consisting of GCN nanosheets (CN)/VO and cetyltrimethylammonium ammonium (CTAB) modified pore-rich GCN nanosheets CCN/VO(Liang et al., 2021). The CN and CCN nanosheets exhibited a sheetlike morphology with a distinctive curling appearance. This composite demonstrated outstanding characteristics, including a higher surface area 500 m<sup>2</sup>g<sup>-1</sup>, exceptional cycle stability of 90.2% over 5000 charge/discharge cycles, and a phenomenal energy density of 60 whkg<sup>-1</sup> at the 1.5 V voltage range. One of the most significant advantages of this composite structure is the increase in the quantity of active sites and surface area. This enhancement results in improved electrical conductivity, reduced charge transfer resistance and enhanced specific capacitance and rate capability. Moreover, these structures contribute to the overall flexibility of supercapacitors by creating suitable pores within the electrode. Additionally, they play a crucial role in enhancing cycle stability during repeated charging and discharging processes. These findings underscore the potential of these composite structures in advancing the performance and stability of supercapacitors, offering valuable insights for the development of high-performance energy storage systems. The increase in cyclic stability and flexibility parameters in 2D structures stands out as a distinguishing feature that has shown comparatively less improvement in 1D structures. Consequently, the 2D structure of GCN emerges as an ideal candidate for fabricating electrodes in supercapacitors. With its numerous advantages, the 2D GCN structure exhibits significant potential and suitability for the development of high-performance supercapacitor electrodes.

Various methods have been designed for synthesizing 2D GCN nanosheets. It is crucial to overcome the van der Waals forces between the carbon nitride layers to produce materials based on 2D GCN. Additionally, the physicochemical characteristics of GCN nanosheets slightly vary depending on the synthesis techniques employed. Therefore, the key to the widespread usage of GCN nanosheets lies in the development of appropriate synthesis procedures tailored to specific application requirements. This emphasizes the importance of continuous research and innovation in refining synthesis approaches to fully unleash the potential of GCN nanosheets across diverse applications. In the first place, it is crucial to highlight that GCN exhibits weak dispersibility in most solvents, which hinders its potential for wider applications. Additionally, the conventional liquid-phase ultrasonic stripping approach, even with a 16-hour ultrasound treatment, only achieves a low concentration of GCN nanosheets, measuring as little as 0.15 mgmL<sup>-1</sup>. This illustrates the limited stripping efficiency associated with the conventional method. These factors emphasize the pressing need for alternative approaches to enhance the dispersibility and stripping efficiency of GCN nanosheets, thereby unlocking their full potential for diverse applications. Newer methods, such as the exfoliation method and self-assembly method, have demonstrated remarkable efficiencies sand hold great potential for advancing the development of 2D GCN materials. For instance, the exfoliation method, specifically gas exfoliation, has proven to be highly effective in fabricating GCN nanosheets, as exemplified by the work of Huayan Si et al (Dong et al., 2013; Si et al., 2020). In their study, bulk GCN was prepared by heating urea in ambient air using a muffle furnace. Subsequently, through the process of exfoliation using liquid nitrogen gasification, GCN nanosheets were successfully

obtained. Remarkably, these nanosheets exhibited a significantly larger surface  $343 \text{ cm}^2 \text{g}^{-1}$  compared to the surface area of bulk GCN, which measured only 72 m<sup>2</sup>g<sup>-1</sup>. This outcome strongly supports the claim that liquid nitrogen gasification is an effective method for exfoliating GCN and obtaining nanosheets with enhanced surface area. These advancements in exfoliation techniques, particularly through the utilization of gas exfoliation, offer a promising pathway for the development of 2D GCN materials with superior properties and expanded functionalities.

An alternative and successful method for obtaining a few-layer GCN is acid-assisted, in-situ exfoliation (Figure 6). By employing this technique, thin porous GCN nanosheets were synthesized, exhibiting a significant surface area of 212.5  $m^2g^{-1}$  and an exceptionally high pore size of 1.55 cm<sup>3</sup>g<sup>-1</sup> (Ong et al., 2016). These porous structures play a dual role by facilitating reactant diffusion and serving as interconnected, extremely thin nanowalls and curved surfaces that effectively function as electron transfer channels within 2D structures. Furthermore, it has been previously mentioned that the presence of a porous structure contributes to the increased structural stability of GCN during the charging and discharging processes of devices Figure 7. These findings underscore the manifold advantages of the acid-assisted, in-situ exfoliation method, enabling the production of a few-layer GCN with enhanced surface area, pore size, and structural stability. Consequently, this method opens up new avenues for the utilization of GCN in diverse fields. Thanks to the enhanced structural modification and inherent doping characteristics, the sample synthesized from inorganic acid protonated melamine (referred to as NCN) exhibited a remarkable reduction of two orders of magnitude in charge transfer resistance (Rct) compared to bulk GCN. This substantial improvement in Rct emphasizes the beneficial properties and potential applications of NCN, underscoring the significance of structural modifications and doping strategies in optimizing the performance of materials based on GCN.



Figure 6. The fabrication of porous ultra-thin GCN nanosheets



# Figure 7. Diverse functions of surface area and pore capacity

Gas bubbling exfoliation has emerged as another innovative method for achieving highly desirable surface characteristics and porosity. A notable example is the work of Hamza Majeed Ansari et al., who reported a gas-bubbling exfoliation strategy assisted by NH<sub>4</sub>Cl to produce ultra-thin 2D GCN nanosheets. Through this approach, the specific surface area of 2D GCN was significantly enhanced to 176.4 m<sup>2</sup>g<sup>-1</sup> (11.6% greater than the reference value for GCN), allowing for a larger water/GCN interface for efficient photo-catalytic water reduction reactions. Furthermore, the increased specific surface area positively impacts the performance parameters of supercapacitors, making this method suitable for structural modification of GCN as an electrode in such devices. Despite the numerous advantages, it is important to acknowledge the limitations of the exfoliation method. These include the time-consuming process, excessive solvent usage, and the potential use of toxic solvents in some methods. Nevertheless, the remarkable achievements and potential applications of gas-bubbling exfoliation underscore its significance as an effective technique for tailoring the properties and performance of GCN materials. Liquid exfoliation is an effective approach that addresses the issue of using toxic solvents by utilizing water as a solvent. In this method, bulk GCN is mixed with a suitable liquid and subjected to ultrasonic processing to achieve exfoliation of GCN (R. Li et al., 2019; Wang, 2019; Yuan et al., 2019). Yao Liu et al. successfully fabricated GCN nanosheets through the liquid exfoliation method(Liu et al., 2020). These nanosheets demonstrated a higher specific surface area compared to bulk GCN. Another effective method is the self-assembled approach, which has received significant attention. For instance, Xiao et al. employed melamine and cyanuric acid to create supramolecular precursors. They further investigated the impact of adding various alcohols to generate 2D GCN nanosheets (Xiao et al., 2019). The study revealed that a solution of ethanol and glycerol can be used as an intercalator, facilitating the layering of GCN into 2D nanosheets. This suggests that polyol, in conjunction with ethanol, can assist in the peeling process of GCN.

In general, all the synthesis methods explored thus far in the field of 2D-GCN have their respective advantages and disadvantages. An ideal method that encompasses all desirable

characteristics has yet to be discovered, and therefore, research and development in this area persist.

#### 3.1.4. 3D structures

3D structures are among the most distinctive and novel architectural forms that different materials can possess. Additionally, the performance of electrodes derived from such structures is influenced by the need for electrochemically inert additives and binders in the design of graphene-based electrodes. Moreover, due to the inherent difficulties associated with the 2D nature of graphene, incorporating additional pseudocapacitive materials onto graphene sheets becomes necessary. To overcome these limitations, the adoption of a 3D structure emerges as an ideal choice. Large surface area 3D porous carbon structures serve as an excellent platform for accommodating active materials. Furthermore, they provide clearly defined pathways to the electrolyte, facilitating efficient charge storage, and ensuring mechanical stability. By incorporating a 3D structure, it becomes possible to leverage these advantages, leading to improved electrode performance, enhanced interactions with the electrolyte, and optimized charge storage capabilities. Therefore, the integration of 3D structures presents a promising pathway for addressing the challenges associated with electrode design, ultimately driving advancements in energy storage systems(Jiang et al., 2018; Kumar et al., 2021).

GCN, being a carbon-based material, also shares the advantages attributed to the 3D structure of carbon-based materials. In addition to its distinctive shape, 3D GCN possesses exceptional electrical, optical, and magnetic capabilities(Duan et al., 2019). Furthermore, when utilized as a binder-free supporting platform for electrode materials in supercapacitors, it exhibits high specific capacitance, reliable rate capability, and excellent cycling stability. The absence of a binder in the synthesis of this material leads to a decrease in charge transfer resistance and an increase in ionic conductivity. These combined parameters render the 3D GCN structure an attractive option for supercapacitor electrode applications. By harnessing the unique properties of 3D GCN, researchers can achieve enhanced performance and effectively address the challenges associated with supercapacitor electrode materials. The intimate connection between material microstructure and performance has propelled the emergence of 3D-ordered nanostructures as a frontier and a highly intriguing research area in nanomaterials. The capability to design and engineer nanostructures in a 3D fashion has unveiled novel prospects for customizing material properties and optimizing overall performance. Through precise control over the arrangement and organization of nanostructures, researchers can unlock enhanced functionalities and achieve improved performance characteristics. The pursuit of 3D-ordered nanostructures represents a captivating pathway toward advancing the field of nanomaterials and exploring their potential applications across various industries.

Extensive research has determined the existence of two primary methods for creating a 3D GCN composite: self-assembly and embedding. The self-assembly method utilizes techniques such as photopolymerization, heating-cold polymerization, and hydrothermal/solvothermal methods. Conversely, the embedding method employs freeze-drying, heat polymerization, and template methods(X. Li et al., 2019). These methods have been thoroughly studied and offer distinct advantages in terms of property customization and performance

enhancement. Researchers can select the most suitable method based on their specific objectives and desired composite properties, thereby expanding the possibilities for fabricating 3D GCN composites. To maintain performance parameters such as cycling stability, specific capacity, energy density, and power density in supercapacitors, it is essential to synthesize a 3D structure for GCN. New structures like flower species, coreshell configurations, or a combination of nano-plate and nanoneedle designs offer promising and less-explored options for GCN. For instance, Dong Liu et al. created a 3D porous GCN with 0D-Fe<sub>2</sub>O<sub>3</sub> to enhance photo-Fenton performance, resulting in increased surface area and active site count. These parameters are crucial for supercapacitor electrode operation, making this type of structure suitable for supercapacitor electrode applications.

The flower-like structure, with its appealing 3D arrangement and ideal properties, is an attractive option for supercapacitor electrodes. Its unique morphology and properties enhance performance by increasing surface area, facilitating ion improving electrochemical and transport, activity. Incorporating the flower-like structure in Supercapacitors improves energy storage capabilities and device efficiency. Duan et al. achieved the synthesis of flower-like 3D GCN structures(Liang et al. 2021). This particular structure demonstrates a significant BET surface area, a rich porous structure, and a wide reaction range for visible light. Experimental findings revealed that the hollow internal structure of the flower-like GCN comprises porous nanosheets, which are likely formed through gas production during the polymerization of MCA. These compelling results provide valuable insights into the formation mechanism and highlight the exceptional properties of the flower-like GCN structure. In another research endeavor, as shown in Figure8 Sajid Ali Ansari and colleagues developed flower-like 3D-SnS<sub>2</sub>/GCN sheets as a promising material for electrochemical supercapacitors(Ansari & Cho, 2017). During discharge, the GCN AP exhibits specific capacitance of 106.06 Fg<sup>-1</sup> and 26.74 Fg<sup>-1</sup> at current densities of 1 Ag<sup>-1</sup> and 5 Ag<sup>-1</sup>, respectively. Correspondingly, the SnS<sub>2</sub>/GCN-pm heterostructure achieved specific capacitances of  $78.83 \text{ Fg}^{-1}$ , 98 Fg<sup>-1</sup> at comparable current loads. Notably, the SnS<sub>2</sub>/GCNst heterostructure surpassed both the GCN-AP and SnS<sub>2</sub>/GCNpm heterostructures in terms of electrochemical capacitance performance, exhibiting specific capacitances of 210.30 Fg<sup>-1</sup> at comparable current loads, both at low and high currents. Furthermore, to assess the potential of the SnS2/GCN-st heterostructures as a durable and stable electrode material, galvanostatic charge and discharge experiments were repeatedly performed on the structure for over 1500 cycles. The tests were conducted at a current load ranging from 0 to 0.4 V of potential, with current of 1 Ag<sup>-1</sup>. While the specific capacitance of the SnS<sub>2</sub>/GCN-st heterostructure exhibited a slight decline with increasing cycle numbers, it maintained a remarkable cycle stability of 84% after 1500 cycles. These findings underscore the exceptional durability and suitability of the SnS<sub>2</sub>/GCN-st heterostructure as a long-lasting electrode Furthermore, electrodes material for Supercapacitors. composed of other materials exhibited a rapid decrease in specific capacitance compared to their initial values. The enhanced cycle stability of the SnS<sub>2</sub>/GCN-St heterostructure electrode can be attributed to its unique 3D design, incorporating two GCN sheets and SnS<sub>2</sub> heterostructures. Additionally, the layered structure of GCN plays a crucial role in reducing interlayer resistance and improving kinetics on the electrode surface. The results demonstrate the significant effect of this structural configuration on increasing the specific surface area and enhancing the porosity of the electrical device. The combination of a nano-plate and nano-needle structure presents an enticing opportunity to create an electrode material with ideal properties for supercapacitors. In an impressive achievement, Xuezhao Wang et al. fabricated a CONi<sub>x</sub>S<sub>y</sub>/GCN composite, which ingeniously integrates the nano-plate structure of GCN with the nano-needle structure of CONi<sub>x</sub>S<sub>y</sub>, resulting in the formation of permeable 3D microspheres resembling an urchin(Wang et al., 2022). Notably, these composite electrodes exhibited an exceptionally high specific capacity of 1029 Fg<sup>-1</sup>, indicating their remarkable electrochemical performance. Moreover, the researchers developed asymmetric Supercapacitors using these composite electrodes, showcasing their potential for achieving high energy density. These Supercapacitors demonstrated an impressive energy density of 71.9 whkg<sup>-1</sup> and exhibited notable cycle stability, retaining 72.2% of their initial capacity even after 5000 cycles. These findings underscore the promising prospects of the CONixSy/GCN composite as an advanced electrode material, offering both high energy storage capacity and long-term stability for Supercapacitors. In a separate research study, Wang et al. delved into the influence of incorporating an ionic liquid and a solvent into an assembly on its behavior(Zhao et al., 2018). The experimental outcomes revealed that the addition of these components resulted in the formation of stable 3D GCN spheres, particularly exhibiting stability even at low concentrations of the ionic liquid. This investigation sheds light on the intriguing effects of

This investigation sheds light on the intriguing effects of incorporating an ionic liquid and a solvent, providing valuable insights into the behavior of the assembled structures. Synthesizing three-dimensional (3D) structures involves more complex synthesis steps compared to their two-dimensional counterparts. However, the advantages provided by 3D structures, such as core-shell and flower-like configurations, include higher specific capacity, improved cyclic stability, and enhanced conductivity for GCN. Therefore, it is crucial to further explore more accessible synthesis methods, such as liquid exfoliation, hydrothermal, solvothermal, and others, for synthesizing GCN. By focusing on these methods, we can fully leverage the advantages offered by 3D structures and harness their strengths to advance the field.



Figure 8. Figure 8. Schematic of the selfassembled 3D GCN made of mixed alcohol intercalated melamine

In general, multi-dimensional structures exhibit improved electrochemical parameters compared to the dimensionless

form of graphene carbon nitride. Among different multidimensional structures, 3-dimensional, 2-dimensional, and 1dimensional structures respectively demonstrate superior electrochemical values about one another. This improvement arises from the increased conductivity, charge transfer speed, and cyclic stability resulting from the larger specific surface area and presence of electrochemically active sites in 3D structures compared to 2D structures, and in 2D structures compared to 1D structures. However, as seen in Figure 9, despite their advantages, multi-dimensional structures still face certain challenges, such as complex, time-consuming, and costly production processes.



### Figure9. Schematic comparison of electrochemical parameters of GCN in 1, 2, and 3- dimensional states with the hardness of its synthesis

These limitations have constrained the progress of GCN with multi-dimensional structures. Consequently, research efforts continue to explore these types of structures and refine their fabrication methods. This paper investigates novel and impactful achievements, including the feasibility of employing non-toxic and environmentally friendly solvents such as water in the process of liquid exfoliation. Additionally, it examines the influence of various solvents and their concentrations on the final properties of the synthesized product. These advancements aim to address the challenges associated with multi-dimensional structures and foster the development of sustainable and efficient production techniques. In analogous review articles, such as the review article authored by Rajat Arora et al. (Arora et al., 2023). Within the purview of this review article, exclusive attention has been directed towards the exploration of the two-dimensional (2D) configuration of GCN or in other study that was done by M.G. Ashritha (Ashritha & Hareesh, 2020). This article solely focuses on the electrochemical parameters of the composites formed by graphene carbon nitride with metal base materials. However, it does not address the discussion of structural features that can potentially enhance the performance of graphene carbon nitride. These similar articles have demonstrated that this review article, apart from addressing a highly pioneering aspect in the field of graphene carbon nitride utilization, has also presented comprehensive information, effectively identifying and addressing existing research gaps. The synthesis methodologies advocated in this comprehensive review article, including melamine self-assembly, liquid exfoliation, among others, have the potential to partially ameliorate the challenges associated with industrialization, such as the exorbitant synthesis costs, by utilizing simpler solvents such as water and adopting easier synthesis routes. However, the primary concern at present has been the synthesis time. While these methods have to some extent reduced this time, further investigations

and experiments in practice are required to minimize the synthesis time as much as possible for industrial production. <u>Table 1</u> presents a compilation of the most recent researches conducted on various nanostructured composite compounds. The results of these researches have further confirmed the statements put forth in our own research. The inclusion of these research findings serves to reinforce the validity and reliability of the claims posited within our study.

#### **4. FUTURE PROSPECT**

In terms of future prospects within the realm of GCN as a supercapacitor electrode, significant advancements are expected to unfold in the not-too-distant future. The immense potential of this material is poised to drive extensive research efforts in this particular field. However, it is essential to acknowledge some of the primary challenges identified through investigations, which holds significant importance for further advancements in this area these primary challenges are: (I) Increasing electrical conductivity and specific capacity of GCN. This case can be improved by creating practical synthesis of different nanostructures or creating different composites. (II) Increasing the efficiency of synthesis methods for the costeffectiveness of GCN production in industrial comparison. Practical examination of new synthesis methods like melamine self-assembly, liquid exfoliation, etc., can be effective in this case. (III) Investigation of innovative nanostructures such as core-shell architectures, sandwich-like structures, flower structures, nano needles, etc., These aforementioned instances hold paramount importance as prospective areas of focus for researchers, as they represent significant perspectives and forthcoming challenges in the utilization of graphene carbon nitride as a supercapacitor electrode.

#### **5. CONCLUSION**

In conclusion, this review article shed light on the importance of energy storage in the face of limited energy resources and a growing global population. Supercapacitors have emerged as a promising solution due to their unique features such as rapid charging and discharging capabilities, structural stability, and high-power density. To address the existing gaps in knowledge, extensive research was conducted to explore the strengths and weaknesses of supercapacitors. Among the various materials investigated, Graphene Carbon Nitride (GCN) has emerged as a new and promising carbon-based material for supercapacitor

electrodes. Despite limited research conducted on GCN as a supercapacitor electrode, this review article has provided valuable insights into its electrochemical properties. The primary objective of this article was to present new research opportunities for synthesizing GCN with enhanced electrochemical properties, thereby inspiring researchers in the field. The content of this review article is the culmination of numerous studies, which have been summarized to present a comprehensive overview of the strengths and weaknesses of 1D, 2D, and 3D-dimensional GCN nanostructures as supercapacitor electrodes. Furthermore, this article has introduced challenges and opportunities in each research area, fostering innovation and encouraging further exploration. By delving into the electrochemical properties of GCN and highlighting potential research directions, this review article aims to unlock the maximum potential of GCN as a supercapacitor electrode. We hope that this article serves as a guiding light for researchers, inspiring them to engage in innovative research endeavors and realize the untapped possibilities offered by GCN. In summary, this review article has contributed to the existing knowledge base by providing a comprehensive evaluation of GCN as a supercapacitor electrode. Its innovative approach, encompassing the strengths of GCN nanostructures in different and weaknesses dimensions, along with the introduction of research challenges and opportunities, sets it apart from previous works. Moving forward, we anticipate that researchers will build upon the findings presented here, driving further advancements in the field of energy storage and supercapacitor technology. Nevertheless, to facilitate industrial advancement in this domain, it is imperative to conduct further investigation into factors such as synthesis time, costs, and complexity. This will enable the identification of promising methods, as highlighted in this review article, aimed at addressing these challenges. Other scholarly articles need to align their focus in a similar direction. Furthermore, this article extends an invitation to researchers and industrialists to aid in the development of this field by engaging with the content, applying the presented solutions to address industrial challenges, and conducting empirical evaluations to compare the efficacy of these solutions in an industrial context.

Nano composite	Specific capacitance	Current	Ref
NCP/GCN	2000F.g <sup>-1</sup>	1Ag <sup>-1</sup>	( <u>Pallavolu et al., 2023</u> )
MnMoO <sub>4</sub> / GCN/CNT	252F.g <sup>-1</sup>	1Ag <sup>-1</sup>	( <u>Rangaraj et al., 2023</u> )
Bi <sub>2</sub> S <sub>3</sub> /GCN	1880Cg <sup>-1</sup>	1Ag <sup>-1</sup>	(Maruthasalamoorthy et al., 2023)
GCN/PBA/NF	3336mFcm <sup>-2</sup>	2mAcm <sup>-</sup> <sub>2</sub>	( <u>Zhang et al., 2023</u> )
NiSe/GCN	320mAhg <sup>-1</sup>	1Ag <sup>-1</sup>	( <u>Khaladkar et al., 2023</u> )

Table 1. Functional results of GCN composite nanostructures as supercapacitor electrodes

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