



# Fabrication and improvement in the electrochemical performance of a 2D g-C<sub>3</sub>N<sub>4</sub>-based symmetric supercapacitor device

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## ABSTRACT

This work evaluates the possibility of employing 2-dimensional graphitic carbon nitride (2D g-C<sub>3</sub>N<sub>4</sub>) to be utilized in supercapacitor applications as an anode material. Compared to traditional carbon-based materials, g-C<sub>3</sub>N<sub>4</sub> (CN) has various benefits, including higher energy-density and rate-capability. Therefore, we investigated the performance of a CN-based supercapacitor synthesized using an efficient and cost-effective thermal polymerization method. X-ray diffraction (XRD) data investigation indicated hexagonal symmetry composed with space group P-6 m2, indicating CN with no discernible presence of any other phases. The XRD pattern was utilised to calculate the average crystallite size, which was around 2.87 nm. In a three-electrode arrangement, electrochemical studies were performed on the CN electrode. The outcomes of this study revealed the specific capacitance (C<sub>s</sub>) to be around 35.2 F/g at 5 mV/s when measured using cyclic voltammetry (CV) and was 39.9 F/g at 1.0 A/g when calculated by galvanostatic charge-discharge (GCD). Finally, the construction of 2D CN based symmetric supercapacitor device was manifested and its electrochemical performance was investigated. Electrochemical studies of the symmetric supercapacitor device demonstrated a highest cell specific capacitance (C<sub>cell</sub>) of 149.1 F/g using GCD at 0.5 A/g. The symmetric supercapacitor device exhibited an extraordinary E<sub>cell</sub> of 141.8 Wh/kg at a P<sub>cell</sub> of 925 W/kg. Overall, this work thoroughly analyzed the electrochemical features of 2D CN and their symmetric supercapacitor devices, and offered insightful information on its prospective use in the energy storage field, which showed that 2D CN has better electrochemical performance than other similar materials.

## 1. Introduction

Over the last few decades, researchers have focused their attention on alternative energy sources owing to the environmental damage caused by the emission of greenhouse gases from fossil fuels. New energy reserve systems, including batteries, fuel-cells, conventional capacitors, and electrochemical supercapacitors with improved performance, must be developed to manage these energy sources [1]. Supercapacitors are one of the solutions for storing energy among various types of devices accessible for outstanding power density, rapid charging and discharging, long lifespan along with low cost. Based on the energy storage

principles, supercapacitors can be divided in three categories: electrical double-layer capacitors (EDLCs), pseudocapacitors, and hybrid capacitors [2]. The electrolyte-electrode interface is where EDLCs store energy by absorbing and desorbing ions. Pseudocapacitors are dependent on reversible reduction and oxidation processes that occur on the surfaces of the electrodes. By combining the capabilities of EDLCs with pseudocapacitors, hybrid supercapacitors are designed. Pseudocapacitors, particularly, are formed of metal oxide and conductive polymers. Although pseudocapacitors have a large specific capacitance, remarkable stability, and a cheap cost, but they have poor electrical conductivity. This is the real difficulty of using pseudocapacitors as a possible

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supercapacitor electrode material [3]. Furthermore, achieving high power and energy densities is another primary requirement for energy storage technologies. To overcome such drawbacks, the electrochemical characteristics of the electrodes may be enhanced via the creation of novel electrode materials, electrolytes, and separators [4] because the electrode material is the primary component affecting the potency of the supercapacitor. For instance, electrode materials influence the specific capacitance due to their electrical active sites, charge storage mechanisms, and electron, and ion transport pathways [5]. A suitable electrode material should have a greater specific surface area (SSA), be inexpensive, readily available, have good conductivity, and be environmentally acceptable. Therefore, finding a suitable electrode material with excellent capacitance performance for a supercapacitor is essential and challenging. Additionally, carbon-based nanostructures are the most often utilized materials for electrodes because of their remarkable conductivity, enormous surface area, and EDLC behavior. However, there are several disadvantages to using carbon materials as electrodes, such as a low energy density, which has led to the limited usage of these materials in real-world applications [6]. Therefore, the focus of the researchers, in recent years, has been shifted to 2D materials owing to their unique characteristics, including an enormous surface area, excellent thermal conductivity, good cyclability, and long-term stability, making them exemplary for usage in supercapacitor applications [7–11]. 2D materials as electrodes offer promising solutions to some of the issues facing supercapacitors, including cost, safety, environmental impact, energy density, and power density.

Among 2D materials, graphitic carbon nitride ( $g\text{-C}_3\text{N}_4$ ) has piqued the interests of scientists in recent decades owing to its distinct assets and variety of applications, including luminescence, photocatalysis, electrocatalysis, optoelectronics, and energy storage devices [12–15]. The  $g\text{-C}_3\text{N}_4$  is recognized as a 2D semiconducting compound of the carbon nitride family, having a band gap of  $\sim 2.7\text{--}2.9$  eV [16]. The  $g\text{-C}_3\text{N}_4$  is chemically and thermally stable, with several active reaction sites in addition to being an excellent electron acceptor and donor. The  $g\text{-C}_3\text{N}_4$  is ecologically benign as it comprises components widely distributed on Earth and excludes unsafe heavy metals. The  $g\text{-C}_3\text{N}_4$  has numerous advantages regarding the environment, including low toxicity, a lower carbon footprint, and abundant and renewable precursors. These characteristics make it a promising candidate for developing environmentally friendly and durable electrode materials for energy storage devices [17]. The  $g\text{-C}_3\text{N}_4$  comprises layers made up from nitrogen, N along with carbon, C atoms organized in a hexagonal geometry, similar to graphite [18]. The core repeating unit of  $g\text{-C}_3\text{N}_4$  comprises tri-s-triazine rings linked through carbon-nitrogen (C-N) bonds, forming a honeycomb-like structure. Although standard carbon-based materials like graphene, activated carbon, and 2D materials like Mxenes are frequently used in supercapacitors,  $g\text{-C}_3\text{N}_4$  offers certain unique benefits. For example, the large surface area, two-dimensional structure, and noteworthy chemical and thermal stability of  $g\text{-C}_3\text{N}_4$  that makes it a preferable choice for high-performance supercapacitors. Furthermore, nitrogen atoms present in  $g\text{-C}_3\text{N}_4$  lattice may improve its electrical conductivity and pseudocapacitive behavior. Consequently, researchers are fascinated by  $g\text{-C}_3\text{N}_4$  due to its potentiality to fabricate energy storage units [19,20]. So, several research groups [21–24] are working on the  $g\text{-C}_3\text{N}_4$  and their composites to exploit their potential to make greater active reaction sites available for application in energy storage. A. Irshad et al. [25] synthesized  $\text{Ag-NiAl}_2\text{O}_4 @g\text{-C}_3\text{N}_4$  exhibited a high specific capacitance of 768 F/g and excellent energy and power densities of 27 Wh/kg and 187.28 W/kg, respectively. The composite showed remarkable cycling stability with 94.01 % capacitance retention after 6000 cycles and a prolonged discharge time of 519 s. The  $\text{Ag-NiAl}_2\text{O}_4 @g\text{-C}_3\text{N}_4$  composite has a charge transfer resistance of only 0.51  $\Omega$ , showing its feasibility in energy storage applications. Tahir et al. synthesized  $g\text{-CN}$  from melamine and tested its electrochemical characteristics in 0.1 M  $\text{Na}_2\text{SO}_4$  reporting specific capacitance  $\sim 71$  F/g at 0.5 A/g current density [26].

A.M. Elsayed et al. prepared  $g\text{-C}_3\text{N}_4/\text{ZnS-ZnO}$  supercapacitors with specific capacitance and energy density of 15.1 F/g and 1.17 Wh/kg, respectively, alongside cycling stability of nearly 88.6 % after 500 cycles of charging and discharging [27]. The symmetric supercapacitor based on  $g\text{-C}_3\text{N}_4$  mesoporous nanosheets was fabricated by L.G. Ghanem and co-workers. The device exhibited a specific capacitance of 78.08 F/g at a scan rate of 5 mV/s in a 0.5 M  $\text{H}_2\text{SO}_4$  electrolyte [28]. M. Subbiah et al. [16] functionalized a  $\text{C}_3\text{N}_4$  nanosheet by protonation via acid treatment. Hydrochloric acid-treated  $\text{C}_3\text{N}_4$  has a maximum specific capacitance of 761 F/g at 20 A/g. A symmetric device with a KOH/LiI gel-based electrolyte has the highest specific capacitance of 175 F/g at 1 A/g.

This work presents the synthesis of CN using a direct and inexpensive thermal polymerization process for supercapacitor applications. The structural properties of synthesized CN are studied by XRD, while Fourier-transform infrared (FT-IR) spectroscopy is employed to study functional groups of synthesized CN. Thermogravimetric analysis (TGA) was used to determine the thermal stability of CN and its precursor, urea. The surface morphology of CN is studied by high-resolution field emission scanning electron microscopy (HR FESEM) and high-resolution-transmission electron microscope (HR-TEM). The electrochemical characteristics of a CN electrode were analyzed using CV, EIS, and GCD in 1.0 M KOH electrolyte. They demonstrated improved performance, indicating that CN is a suitable material for anodes used in energy storage applications. The symmetric supercapacitor based on CN was fabricated, and its electrochemical characteristics were analyzed in an alkaline electrolyte (1.0 M KOH), making it ideal for energy storage devices.

## 2. Experimental details

The thermal polymerization synthesis method was used to prepare  $g\text{-C}_3\text{N}_4$  [29]. In this method, 5 gm urea (mol.wt.–60.06 g/mol) (Molychem) (99 % pure) was incorporated in a silica crucible which is provided with a cover to circumvent degradation. In a muffle furnace, the material was heated to 550°C at a rate of 5°C per minute for 3 h. The pale-yellow substance was generated and crushed into powder with an agate mortar after reaching room temperature. The  $g\text{-C}_3\text{N}_4$  is named as CN. A sample of CN was obtained as a consequence. The XRD pattern was acquired with the help of the Bruker D8 advanced EcoPro X-Ray Diffractometer ( $\text{Cu K}\alpha$  ( $\lambda = 1.54056$  Å)). A PerkinElmer FT-IR Spectrometer was used to record the FT-IR spectra of the CN. TGA was carried out in an inert nitrogen surrounding and an alumina pan using a thermal analyzer (NEXTA STA 200, Hitachi High-Tech Science Corporation, Japan) with a heating rate of 20 °C/min. The observations were made between 25°C and 900°C in temperature. The surface morphologies were analyzed using a JEOL JSM-7900F FESEM with a 15.0 kV acceleration voltage. The surface area and porosity of CN were studied using Brunauer–Emmett–Teller (BET) testing (Anton Paar, Autosorb 6100 FKM MP-AG). After that, the CN electrode was prepared for anode testing. The electrode was prepared by combining CN, polyvinylidene fluoride (PVDF) (mol.wt.–534000 by GPC) (Sigma Aldrich) and carbon black in an 80:10:10 ratio. N-methyl-2-pyrrolidone (NMP) was subsequently mixed with the combination to produce a slurry solution. The slurry was poured on nickel foam (1.0 cm×0.5 cm) using a drop cast technique. The CN-based electrode was then dried for 12 h at 80°C in a hot air oven. The CN-based electrode was employed as a working electrode in electrochemical tests after drying. Using CV, GCD and EIS, the electrochemical properties of the CN sample were assessed. All electrochemical investigations were carried out with an electrochemical instrument (Biologic VSP-300) and a conventional three-electrode configuration. In a three-electrode system, the CN electrode was used as the working electrode, Pt wire was used as the counter electrode, and Ag/AgCl was used as the reference electrode. A 1.0 M KOH solution is utilized as the electrolyte solution. The applied potential range for the CV studies was +0.0 V vs. Ag/AgCl to +0.8 V vs. Ag/AgCl, whereas the voltage window for the GCD analyses was +0.0 V vs. Ag/AgCl to +0.7 V

vs. Ag/AgCl. The following equation was used to measure the  $C_s$  from CV. [30],

$$C_s = \frac{\int I \cdot dV}{2m \cdot \theta \cdot \Delta V} \quad (1)$$

In which  $\int I \cdot dV$  is an area under the curve from the cycle of CV,  $m$  represents the mass put onto the nickel foam,  $\theta$  is a scan rate,  $\Delta V$  is applied potential difference. The below equation is used to calculate the energy density, power density, and  $C_s$  values of the CN electrode using GCD [30],

$$C_s = \frac{I \cdot \Delta t}{m \cdot \Delta V} \quad (2)$$

$$\text{Energy Density} = \frac{C \Delta V^2}{2} \quad (3)$$

$$\text{Power Density} = \frac{3600 \times E}{\Delta t} \quad (4)$$

Here,  $I$  denote applied current to discharge,  $\Delta t$  is time for discharge,  $m$ , and  $\Delta V$  is as above. The range in which the EIS data were collected was 0.1 Hz to 100 kHz.

The electrode for the CN-based symmetric supercapacitor device was made using the drop-casting method. The electrode was created by blending CN, PVDF, and carbon black in an 80:10:10 ratio. After that, the mixture was mixed with NMP to make a slurry solution. The drop-casting method was used to place the slurry onto a graphitic paper substrate, and the electrode for the CN-based symmetric supercapacitor was created. Subsequently, a hot air oven was used to dry the CN-based symmetric supercapacitor electrode for 12 h at 80°C. The supercapacitor device was developed using Swagelok cells and Whatman filter paper as separators. The electrochemical performance of a symmetric supercapacitor in a two-electrode arrangement with a 1.0 M KOH electrolyte was examined using CV, GCD, and EIS. Fig. 1 demonstrates the schematic of two electrode symmetric supercapacitors. The  $C_{\text{cell}}$  of the symmetric supercapacitor was calculated utilizing the following formula using the CV and GCD curves [31],

$$C_{\text{cell}}(\text{CV}) = \frac{\int I \cdot dV}{m \cdot \theta \cdot \Delta V} \quad (F \cdot g^{-1}) \quad (5)$$

$$C_{\text{cell}}(\text{GCD}) = 2 \times \frac{I \cdot \Delta t}{m \cdot \Delta V} \quad (F \cdot g^{-1}) \quad (6)$$

In which,  $\int I \cdot dV$  is a CV area under the curve,  $m$  denotes the total mass loaded on the substrate,  $\theta$  denotes scan rate,  $\Delta V$  denotes an

electrochemical voltage window, the applied current is represented as  $I$ , and  $\Delta t$  denotes a discharging time. The following equation is used to compute the  $E_{\text{cell}}$  and  $P_{\text{cell}}$  [31].

$$E_{\text{cell}} = \frac{1}{4 \times 3.6} \cdot C_{\text{cell}} \cdot \Delta V^2 \quad (\text{Whkg}^{-1}) \quad (7)$$

$$P_{\text{cell}} = \frac{E}{\Delta t} \times 3600 \quad (\text{Wkg}^{-1}) \quad (8)$$

### 3. Results and discussion

#### 3.1. X-ray diffraction analysis

Fig. 2(a) displays the XRD pattern of CN. It is evident from the XRD pattern that the CN is crystallized [32]. It is noteworthy to discuss that the XRD peaks were observed at 13.10° and 27.30°, which correspond to the (100) plane and the (002) plane, respectively, match the JCPDS card number 87-1526. The in-plane arrangement of heptazine structure is associated with the XRD peak that is given to the (100) plane at approximately 13.10°, whereas the inter-layer stacking of the 1, 3, 5 triazine (melem plane) is related to the diffraction peak that is assigned to the (002) plane at about 27.30°. Eq. (9) is used to determine the interplanar distance ( $d$ ) of the (002) and (100) planes, which was found to be 3.26 Å and 6.75 Å, respectively [33,34].

$$n\lambda = 2d \cdot \sin\theta \quad (9)$$

Where the variable  $n$  represents an integer,  $\lambda$  denotes the X-ray's wavelength, and  $\theta$  represents the angle between the incident X-ray and the scattering plane. The lattice parameters for CN are  $a = b = 4.7720$  Å and  $c = 6.4905$  Å, which were calculated using "powderX" software. These values imply to the hexagonal phase corresponding to the space group P-6m2 (187). The crystalline size of CN is 2.87 nm, which is calculated from the dominant (002) plane peak by the Debye-Scherrer formula (Eq. 10) [35],

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (10)$$

Here,  $D$  represents the crystalline size of the sample,  $K$  is a shape factor constant,  $\beta$  indicates an FWHM of XRD peaks, and  $\theta$  represents the incident angle. No other impurity peak was observed, which confirms the synthesis of pure CN. Our findings are consistent with earlier research by Prakash et al. [36] and Molaei et al. [37], who synthesized g-C<sub>3</sub>N<sub>4</sub> nanosheets with similar features. Molaei et al. reported a 5 nm crystallite size and an effective photocatalyst. Furthermore, Prakash et al. showed effective degradation of RhB and CV dyes in 70 and 60 min, respectively, and attained a crystallite size of 68 nm. In this study, the crystalline size of CN is 2.87 nm. Smaller crystallite sizes improve supercapacitor performance by increasing the SSA and offering more active sites for charge storage. They provide increased power density and faster charge-discharge operations by reducing electron and ion transport pathways. Furthermore, smaller crystallites improve structural stability by absorbing mechanical stresses during cycling and enhancing long-term performance [36,37].

#### 3.2. FT-IR analysis

The FTIR spectrum was analyzed to determine the presence of functional groups. Fig. 2(b) shows the FT-IR spectrum of CN. A prominent peak at 810 cm<sup>-1</sup> in the FTIR spectrum suggests heptazine (tri-s-triazine) unit's bending vibrations in CN. The broad band at 2900–3600 cm<sup>-1</sup> appeared due to stretching vibrations of amino groups containing N-H bonds in the CN. This range also represents the hydroxyl group (O-H) due to adsorbed H<sub>2</sub>O molecule. Further, stretching vibrations of Aromatic heterocycle i.e., C-N and C=N, produce multiple strong peaks in the 1240–1640 cm<sup>-1</sup> range. [38]. No additional

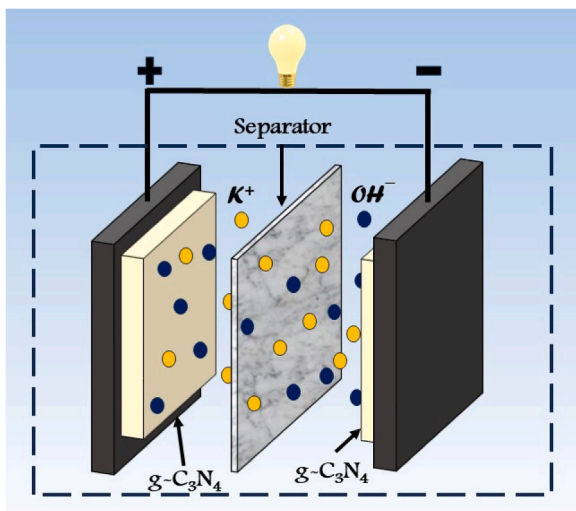


Fig. 1. Schematic diagram of CN-based symmetric supercapacitor.

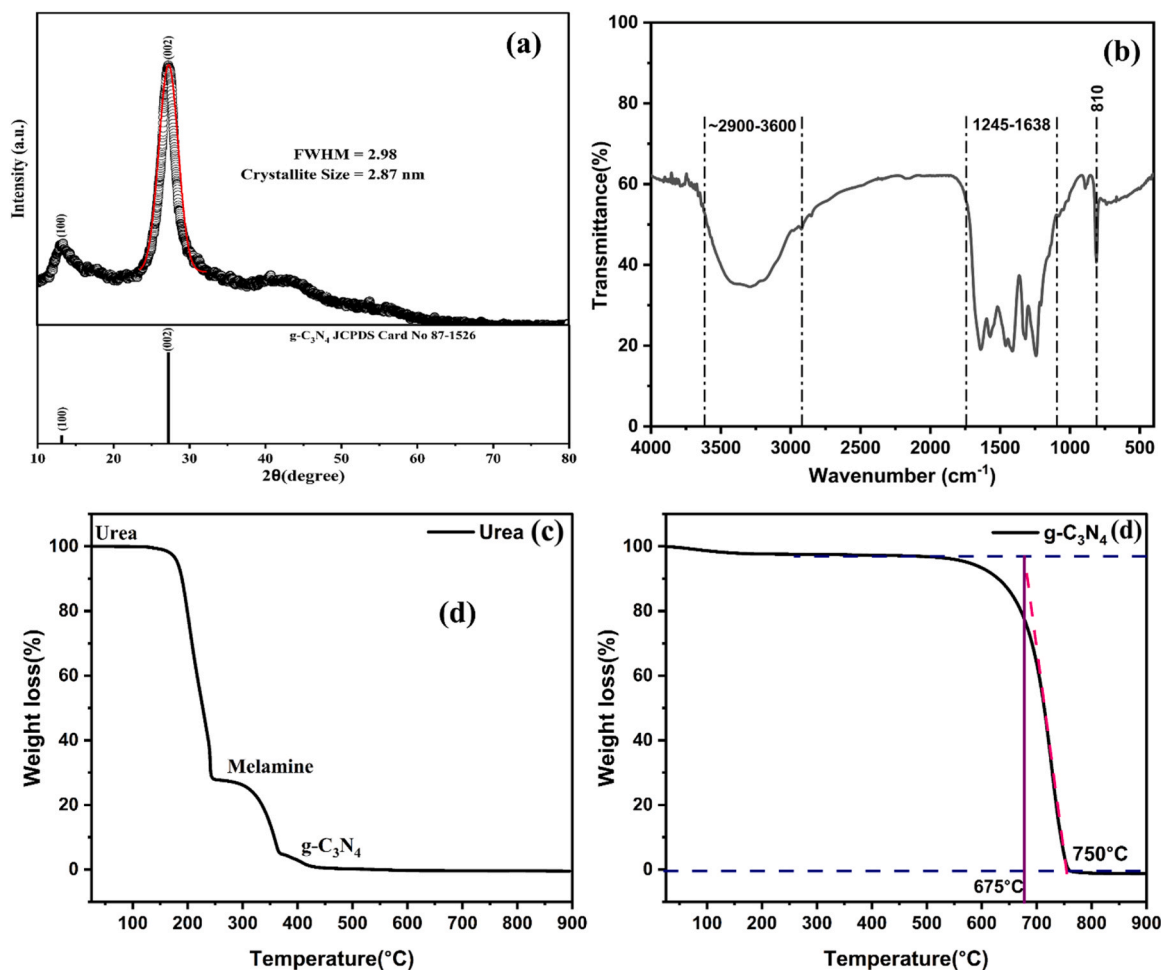


Fig. 2. (a). XRD pattern of CN, (b). FT-IR spectra of CN, Thermogravimetric analyzer curve of (c). Urea, (d). CN.

functional groups are seen, confirming that CN has no other impurities, which is consistent with the XRD results.

### 3.3. Thermogravimetric analysis (TGA)

The TGA was employed to investigate the thermal stability of CN and its precursor (urea). Fig. 2(c) and (d) show the thermogravimetric analysis curve of urea and CN, respectively. The TGA curve was recorded from 25°C to 900°C with a 20 °C/min heating rate. The TGA curve of urea shows a mass drop of about 70 % between 150°C and 250°C, as shown in Fig. 2(c). In this temperature range, urea sublimation and condensation occur, resulting in the formation of melamine and the removal of ammonia [39]. Melamine condensation and sublimation occur between 250°C and 500°C resulting in the synthesis of polymeric carbon nitride via melem and melon as well as the removal of ammonia [40]. Urea should further polymerize to graphitic carbon nitride sheet-type structures at temperatures over 500°C. These structures become unstable at temperatures of about 650°C and release some toxic byproducts of breakdown [41]. Thus, it can be concluded that the temperature of 550°C is more favorable for the thermal polymerization method for the synthesis of CN from urea.

Fig. 2(d) displays a TGA curve of the synthesized CN sample. It is observed that there was just one significant mass decrease (100 %) in the TGA curve of the CN which happened between 500 and 700°C. The decomposition process began slowly at 500°C and weight began to decrease gradually between 550°C and 675°C while after 675°C, the mass decreased drastically. This decomposition occurs when the chemical bond between polymeric melem units in graphitic carbon nitride is

broken at higher temperatures resulting in the emission of gases which include nitrogen, carbon and cyano units [42]. The material decomposes entirely at temperatures up to 750°C leaving minimal residue. As a result, CN exhibits good thermal stability up to 750°C.

### 3.4. FE-SEM analysis

Fig. 3(a) shows FE-SEM images of CN. FESEM analysis revealed a sheet-like structure for the CN samples with numerous pores and a rough surface. This finding suggests that holes were formed throughout the synthesis process as a consequence of urea decomposition. Additionally, the porous and rough structure of CN enables rapid electron transport during electrochemical reactions. The porous and rough structure enhances the capabilities of supercapacitor by maximizing electrochemical characteristics [43]. The element mapping pictures of CN are shown in Fig. 3(b–d). These mapping pictures indicate the presence of elements in the sample: C (in red) and N (in cyan), combined in Fig. 3(b), while Fig. 3 (c–d) displays individual images of N in cyan and C in red, respectively. The energy-dispersive X-ray spectroscopy (EDS) spectrum for the CN is shown in Fig. 3(e). The spectrum validates the formation of the CN by confirming the presence of components C and N. The EDS spectrum shows a Pt peak, suggesting the deposition of a Pt layer to generate a conductive material surface for FE-SEM experiments.

### 3.5. HR-TEM analysis

To learn more about the microstructure and phase information of CN, HR-TEM and SAED investigations were carried out. The HR-TEM

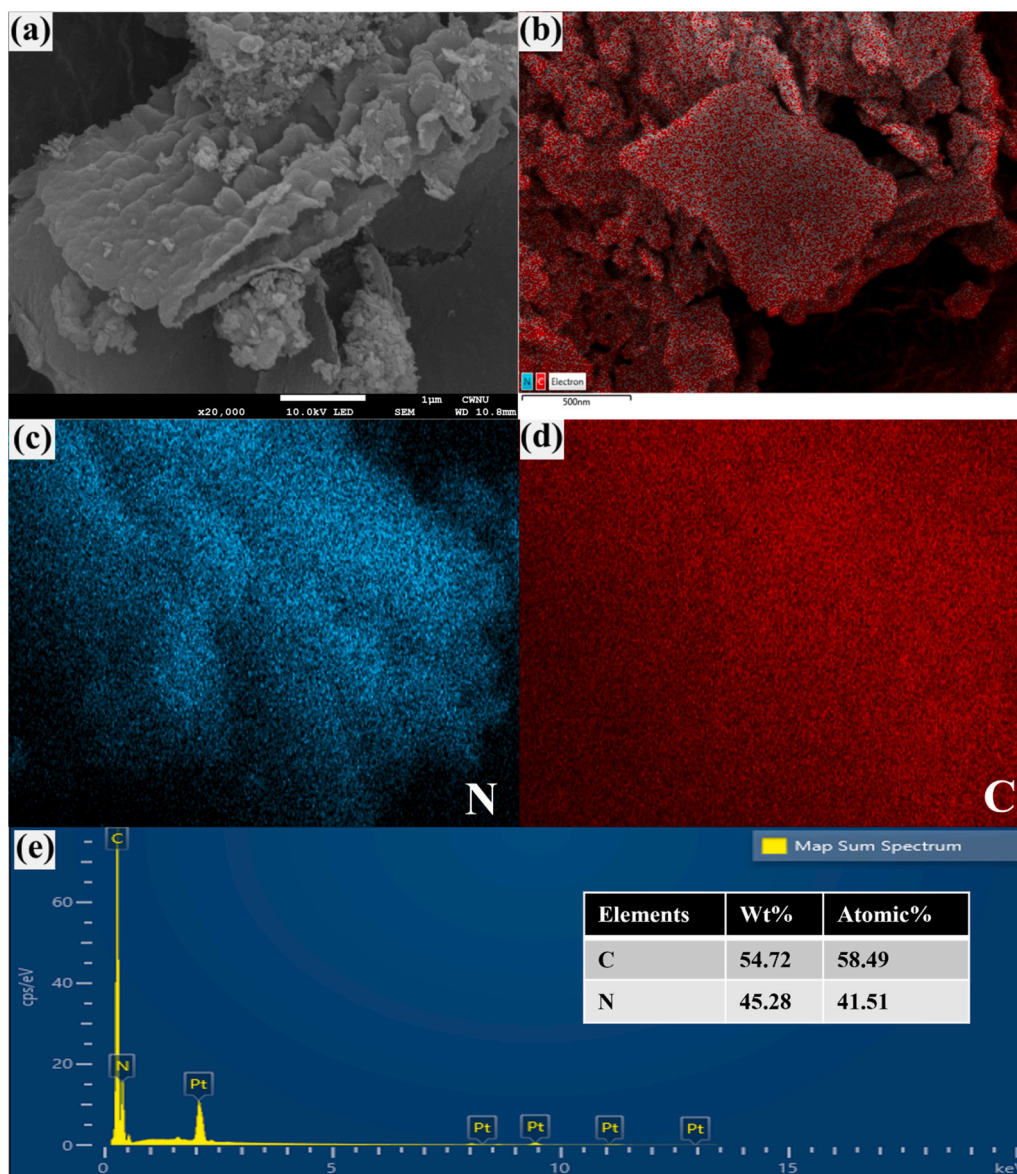


Fig. 3. (a). CN FESEM images at low magnification (b, c & d). elemental mapping of CN, (e). and corresponding EDS spectrum.

micrographs of the prepared sample are shown in Fig. 4(a). The micrographs demonstrate that the nanoparticles have a consistently distributed elliptical shape. The size distribution histogram in Fig. 4(b) shows a wide range distribution, with an average particle size of  $\sim 58$  nm. Additionally, the HR-TEM image of CN is shown in Fig. 4 (Inset), which is used to determine the value of  $d$ . The value of  $d$  determined using an HR-TEM micrograph was 0.33 nm, which matches to the (002) plane of CN. Fig. 4(c) depicts the SAED pattern for CN, featuring one ring (002) that corresponds to reflections with the P-6m2 hexagonal symmetry [44].

### 3.6. BET analysis

The SSA and pore size of CN has been investigated in the context of adsorption isotherms of nitrogen at a temperature of 77 K. Figs. 5(a) and 5(b) illustrate the curves of CN's  $N_2$  adsorption/desorption isotherms and Barrett-Joyner-Halenda (BJH) pore size distribution. The CN sample exhibited a SSA of  $43.2 \text{ m}^2/\text{g}$ , an average pore size of  $37.8 \text{ \AA}$ , and a total pore volume of  $2.64 \text{ cm}^3/\text{g}$ . The CN sample had a significant quantity of mesopores with dimensions ranging from  $29$  to  $50 \text{ \AA}$ , whereas it had a limited amount of macropores with dimensions ranging from  $50$  to  $70 \text{ \AA}$ .

The trace of mesopores and macropores in electrode facilitates the ion-diffusion from the electrolyte which in turn contributes to an increase in the capacitive property of the sample. Based on the results of the BET study, it was determined that CN electrodes had a good SSA and the average pore size, making them more suitable for the diffusion process in supercapacitor applications [42,45,46].

### 3.7. Electronic structure analysis

XPS was used to analyze the sample's oxidation state and chemical composition. Fig. 6(a) displays survey scan spectra of the CN nanosheets which clearly indicate the presence of significant peaks for carbon, nitrogen, and oxygen at  $284.27 \text{ eV}$ ,  $395.27 \text{ eV}$  and  $529.27 \text{ eV}$ , respectively. The presence of oxygen peaks in the broad spectrum is caused by environmental moisture present in the sample. Fig. 6(b) shows the deconvolution C1s spectra with peaks appearing at  $281.29 \text{ eV}$ ,  $284.67 \text{ eV}$  and  $289.09 \text{ eV}$ , respectively, indicating the  $\text{sp}^2$  hybridized graphitic carbon atoms in C-C coordination, the  $\text{sp}^3$  bonded carbon in N-C=N coordination, and the C=O, respectively [47–49]. Fig. 6(c) depicts the deconvolution spectra of the N 1s spectra, with four peaks at  $395.07 \text{ eV}$ ,  $396.02 \text{ eV}$ ,  $397.63 \text{ eV}$ , and  $400.70 \text{ eV}$  corresponding to the

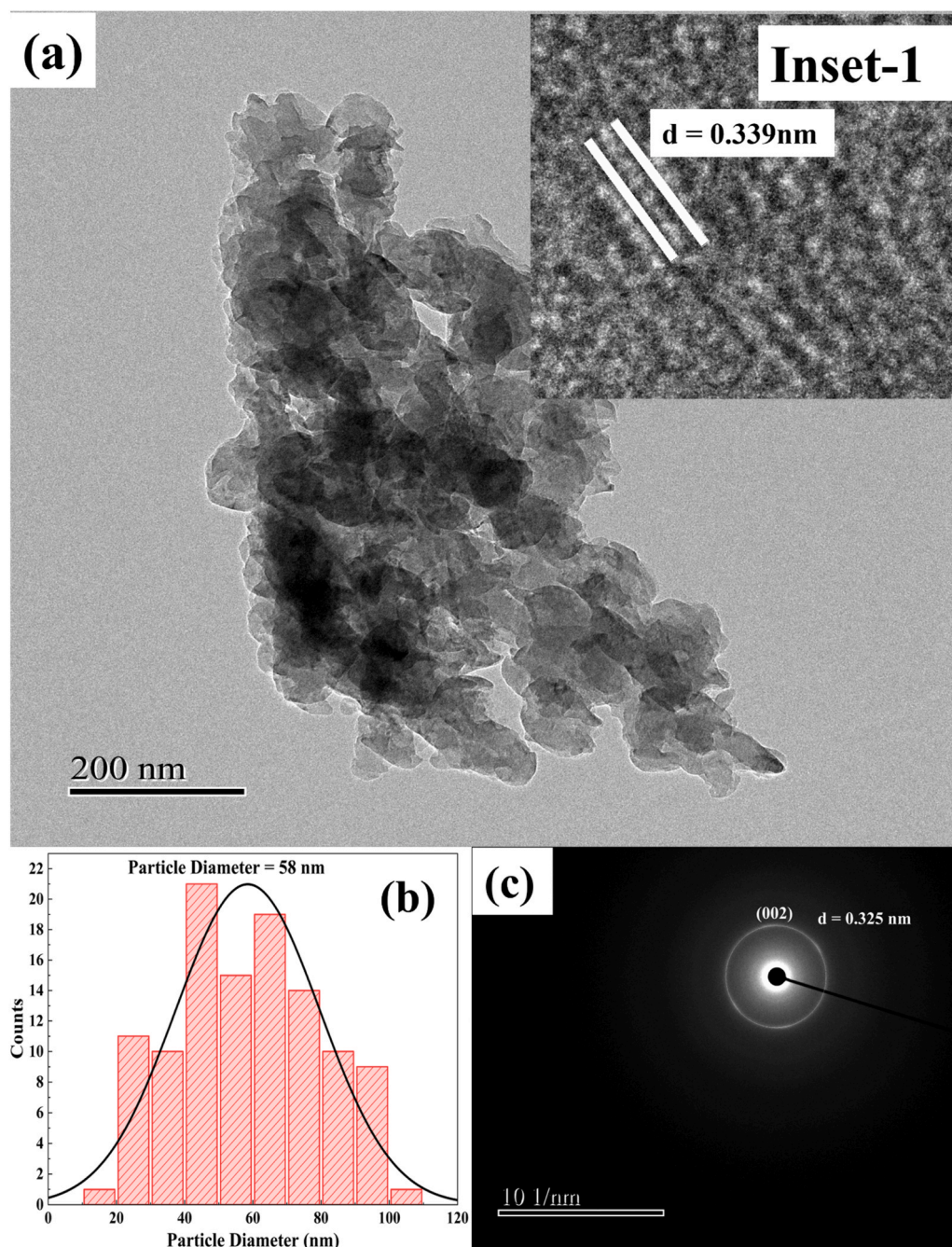


Fig. 4. (a) TEM micrographs and inset-1 highlights HR-TEM micrograph; (b) size distribution histograms; (c) SAED patterns of CN.

C-N, C=N, N<sub>2</sub>-C and C=N-H, respectively [48,50]. The XPS findings give excellent evidence for the chemical composition of the CN, which helps to understand its structural and chemical features, leading to a better understanding of the electrochemical characteristics of CN.

### 3.8. Electrochemical performance of CN electrode in three-electrode setup

The conventional three-electrode system was used to measure the electrode performance in a 1.0 M KOH electrolyte solution. Fig. 7(a) displays a CV graph of the CN-based electrode tests in a voltage range of + 0.0 V to + 0.8 V. The measurements were performed with scan rates varying between 5 mV/s to 100 mV/s. The current response of the CN electrode enhanced as scan rates increased, which amply demonstrated the capacitive behaviour of the CN electrode and was consistent with results that had been previously published [3]. The CV plot of the CN

electrode displayed oxidation and reduction peaks, which abundantly illustrates the verified pseudocapacitive behaviour of the CN [51]. The CV plot shows oxidation and reduction peaks centered at 0.67 V and 0.53 V, respectively. Furthermore, the  $C_s$  of the CN electrode was derived from Eq. (1) using a CV plot. Fig. 7(b) displays a  $C_s$  vs scan rate plot. The  $C_s$  values of the CN electrode were found to 35.2 F/g, 32.5 F/g, 29.8 F/g, 27.5 F/g, 26.2 F/g, 25.4 F/g, 23.3 F/g and 22.0 F/g at a scan rate 5 mV/s, 10 mV/s, 20 mV/s, 30 mV/s, 40 mV/s, 50 mV/s and 100 mV/s, respectively. As the scan rate rises, the  $C_s$  value decreases because, at higher scan rates, the ions of the electrolyte have a shorter amount of time to enter deeply into the pores of the electrode. As a consequence, fewer ions are involved in the charge storage process, which leads to a lower capacitance [52]. At lower scan rates, the ions of electrolyte permeate into the active regions of the electrode increasing the charge storage as well as the  $C_s$  value [3].  $C_s$  declines with increased

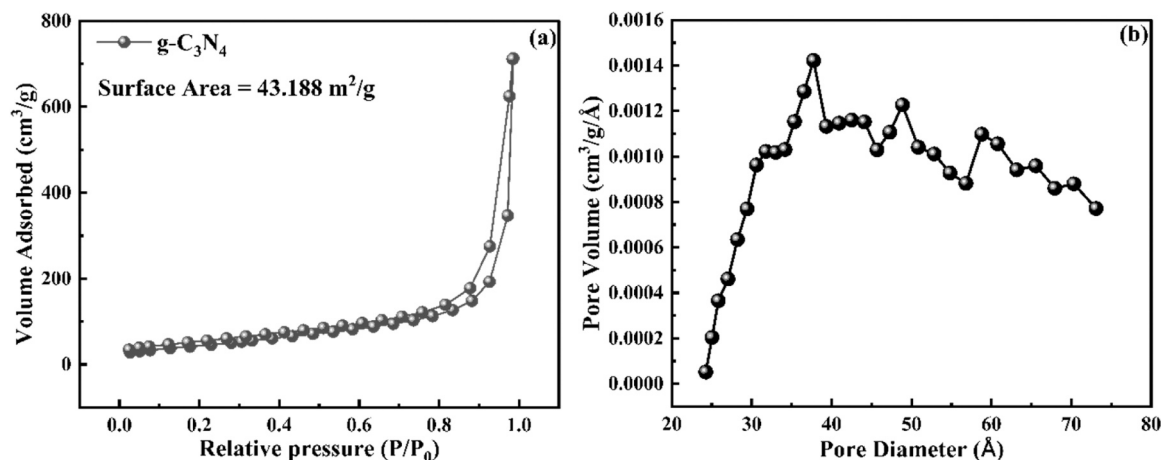


Fig. 5. (a) Surface area isotherms characteristics of CN, (b) Pore size distribution graph of CN.

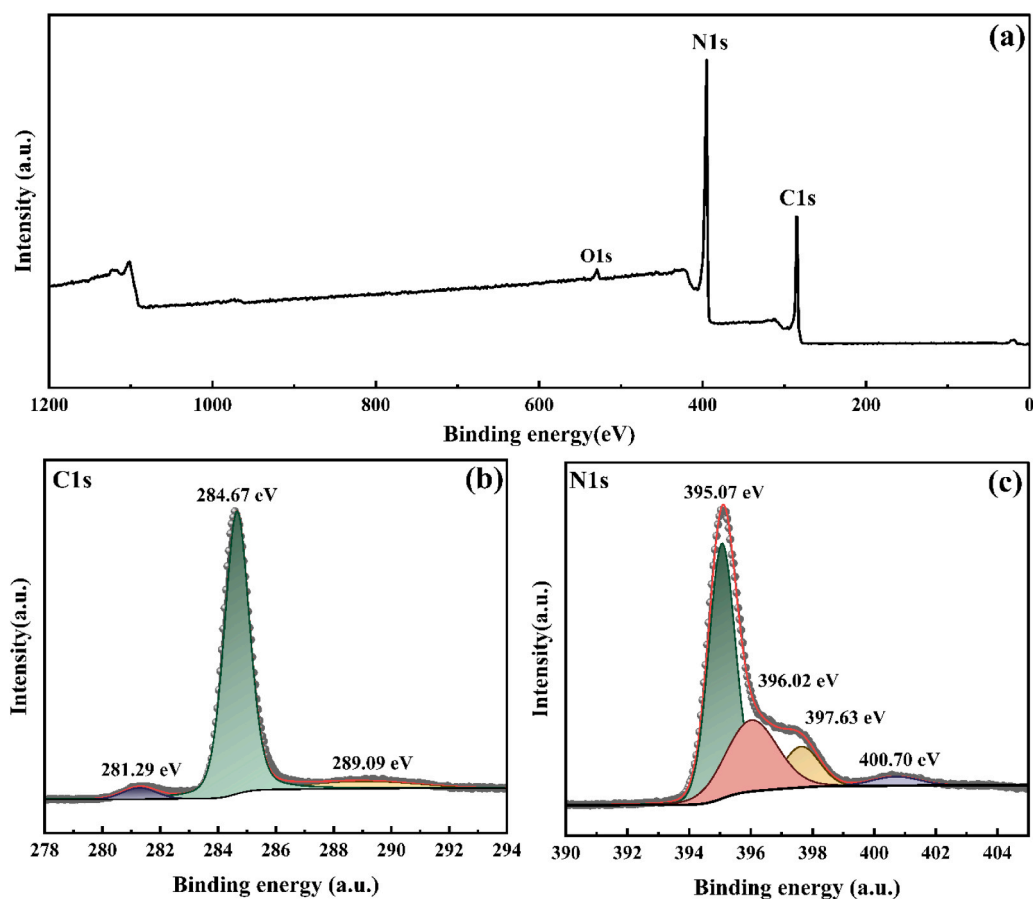
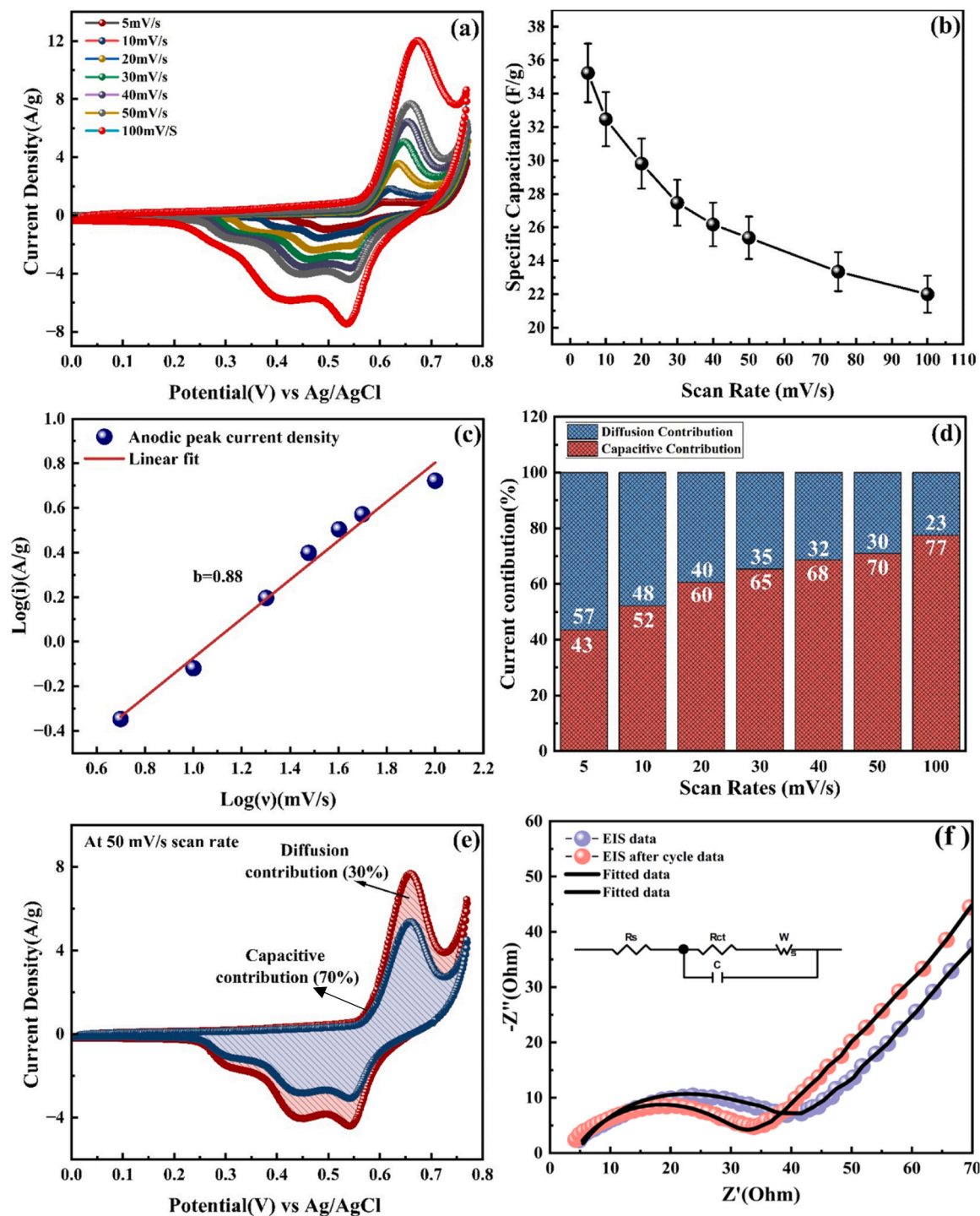


Fig. 6. XPS spectrum of CN, (a), survey spectra, (b). C1s, and (c). N1s.

scan rates, mainly owing to diffusion limitations. This is evident from the analysis of the charge storage mechanism. Reducing electrode thickness may help to improve rate capability and lessen diffusion limitations. Additionally, optimizing the electrolyte solution may improve ion transport kinetics and overall performance [53]. To examine the charge storage mechanism of the CN electrode, Eq. (11) was used to determine the dependence of anodic peak current on the scan rate utilizing the current power law. The logarithmic representation of current density versus scan rate can be observed in Fig. 7(c) [54,55].

$$i = av^b \quad (11)$$

This formula has an anodic peak current ( $i$ ) and a scan rate ( $v$ ), with  $a$  and  $b$  being variables that can be changed. A surface-controlled charge storage mechanism is indicated when the value of  $b$  is  $\sim 1$ , while a diffusion-controlled mechanism is dominated when  $b$  is  $\sim 0.5$ . Fig. 7(c) yielded the  $b$  value of the CN electrode  $\sim 0.88$  which suggested that the primary cause of storing charge in the CN electrode is a surface-controlled mechanism [55]. The total current density ( $i(V)$ ) is determined by the combined contributions of the surface-controlled ( $k_1.v$ ) and diffusion-controlled ( $k_2.v^{1/2}$ ) electrochemical processes. These components were calculated using Eqs. (12) and (13) [56,57].



**Fig. 7.** (a) C-V curve at several scan rates ranging from 5 mV/s to 100 mV/s, (b) Specific capacitance vs scan rates graph of CN electrode, (c) log(i) versus log (v) plot (d) Capacitive and diffusion contribution graphs of CN, (e) A percentage of the CV curve representing the surface and diffusion contributions of CN at 50 mV/s, (f) Nyquist plot of CN electrode from 0.1 Hz to 100 kHz before and after 1000 cycles.

$$\frac{i(V)}{v^{1/2}} = k_1 v^{1/2} + k_2 \quad (12)$$

$$i(V) = k_1 v + k_2 v^{1/2} \quad (13)$$

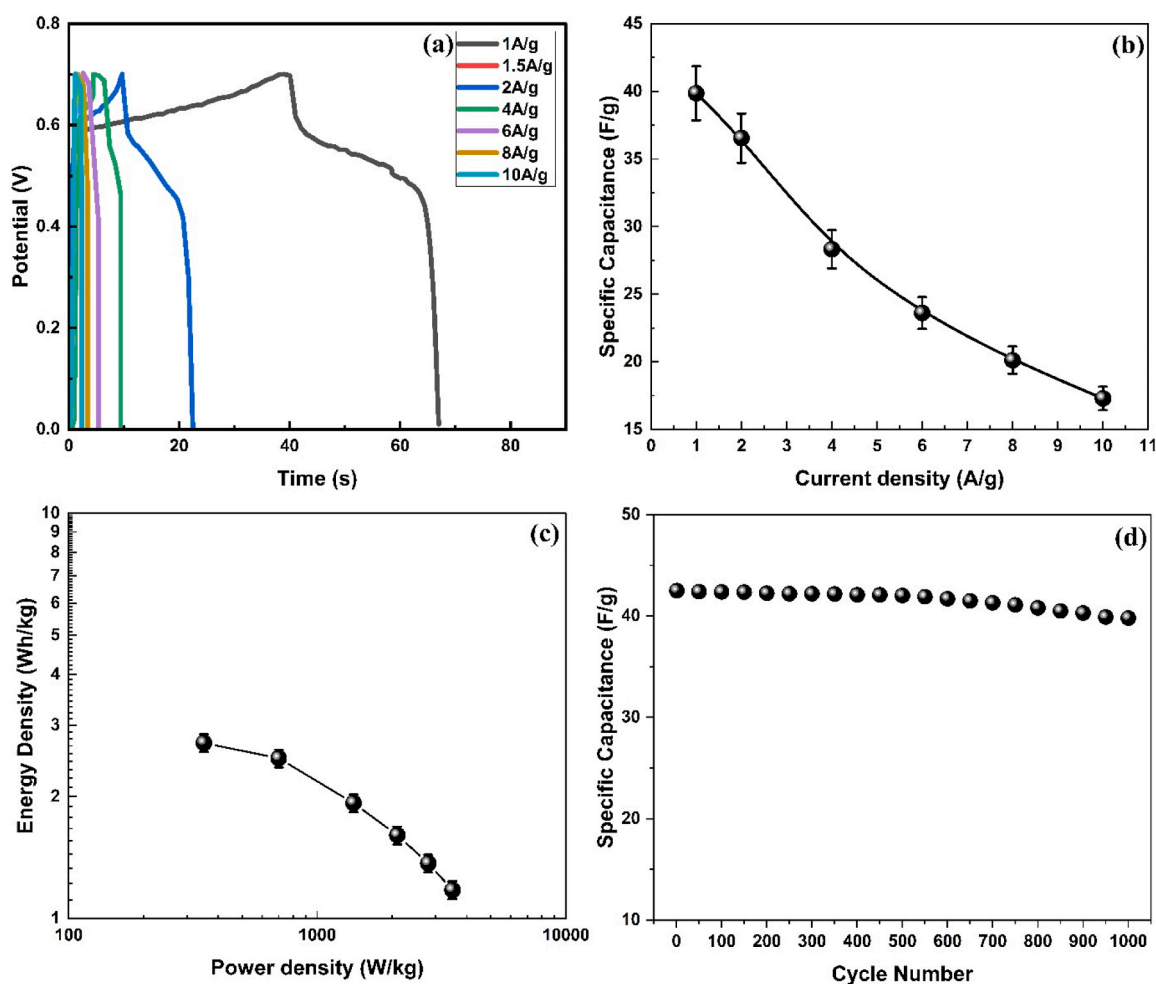
Using these formulas, we can get the intercept of the Y axis ( $k_2$ ) and slope ( $k_1$ ) by plotting  $i(V)/v^{1/2}$  vs.  $v^{1/2}$ . Compared to the total capacity of the CN electrode, the ratio of surface-controlled and diffusion-controlled faradaic mechanisms is shown in Fig. 7(d). At a scan rate of 5 mV/s, the

primary characteristics of the CN electrode are 43 % surface-controlled capacitive mechanisms. Additionally, when the scan rate rises, the diffusion-controlled process's effect diminishes, and surface-controlled redox reactions become prominent, suggesting that electrolyte intercalation into the CN electrode interface occurs [43]. The CV curve for the CN electrode is depicted in Fig. 7(e) at a scan rate of 50 mV/s. This curve displays the current contribution of the electrode which includes both surface-controlled processes (70 %) and diffusion-controlled processes (30 %) resulting in a total contribution of 100 %. EIS experiments are

conducted to examine the transfer of electrolyte ions and the electrical conductivity of the CN electrode. The Nyquist impedance plots for the fabricated CN electrode before and after 1000 charging-discharging cycles measured at open circuit voltage (OCV) and in the frequency span of 1 MHz to 100 mHz as shown in Fig. 7(f). A + 5.0 mV input signal was conveyed to the electrode. The plot depicts the electron transport parameters at the electrolyte and electrode interfaces. The high-frequency section of Nyquist plots is semicircular, whereas the low-frequency region has a linear shape. The low-frequency linear trend reveals Warburg resistance which is the result of frequency-dependent ion diffusion/transport from the electrolyte to the interface of the electrode. The resistance to charge transport on the electrode is represented by the semicircle diameter [58–60]. Fig. 7(f) shows a Nyquist plot of the CN before and after 1000 charging-discharging cycles. The CN electrode exhibits a Warburg resistance of around  $\sim 37.08 \Omega$ , representing a diffusive resistance of hydroxyl ions inside the electrode and exhibiting capacitive characteristics. After 1000 cycles of charging-discharging, Warburg resistance is lowered from 37.08 to  $31.16 \Omega$ . It is due to over-frequent charge-discharge cycles that the electrodes undergo degradation. This deterioration may result in modifications to the electrode surface structure, porosity, or active material characteristics. Such alterations may influence the diffusion properties of ions, influencing the Warburg resistance [61]. The solution resistance ( $R_s$ ) was determined by the intercept of the Nyquist plot on the real axis at high frequencies. This value, which represents the combined intrinsic resistance of the electrode/electrolyte interface and the contact resistance, was found to be lower for CN after 1000 cycles ( $4.02 \Omega$ ) compared

to its initial value of  $4.48 \Omega$  [43]. The resistance of charge transfer ( $R_{ct}$ ) of the CN electrode before and after 1000 charging-discharging cycles are  $33 \Omega$  and  $28 \Omega$ , respectively. It is seen that after 1000 cycles of discharging and charging, the  $R_{ct}$  value of the CN electrode decreases, which may be due to the fast charge transfer in the CN electrodes [62].

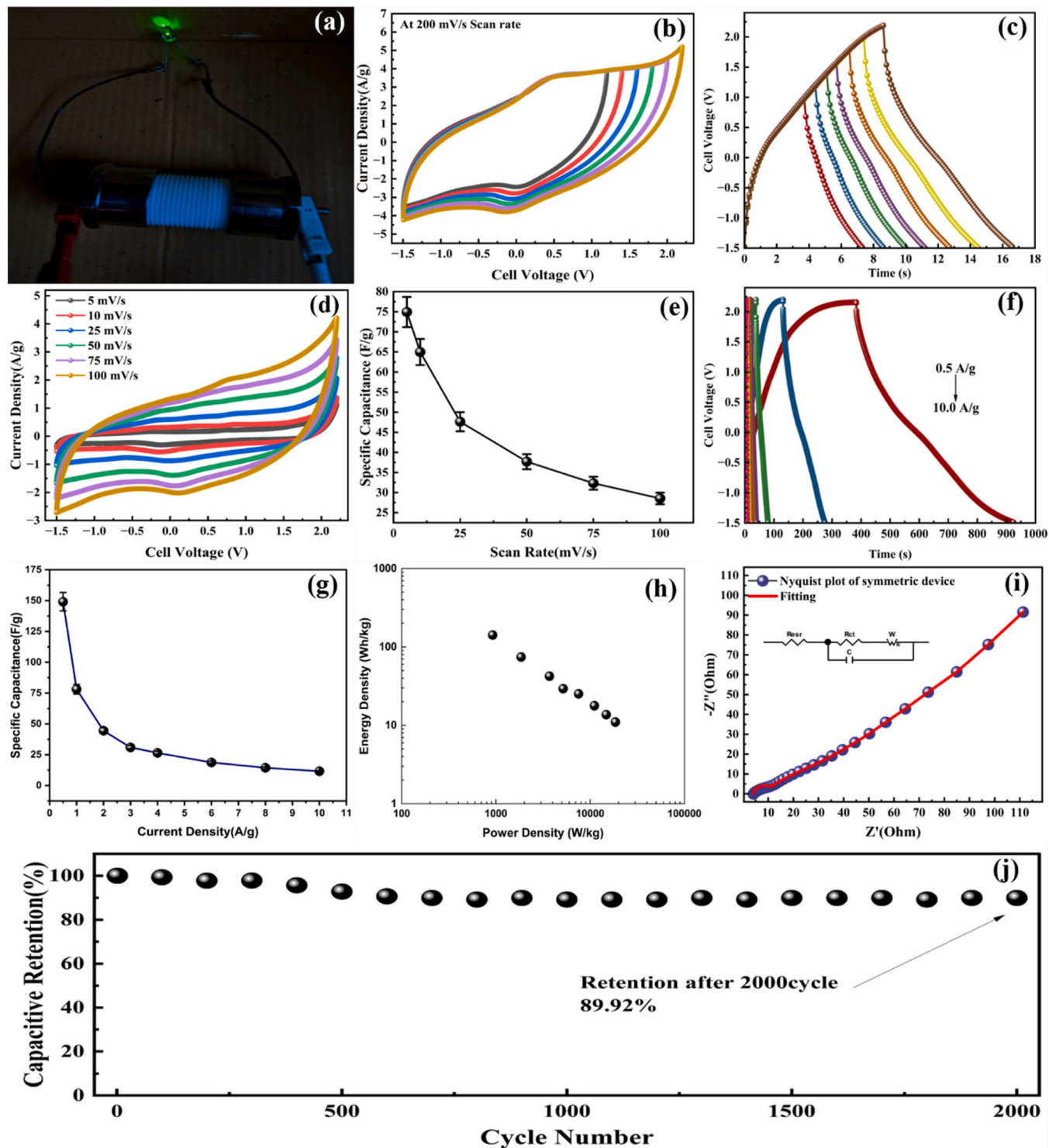
To learn more about the characteristics of the CN electrode, the GCD curves that have been collected at various current densities varying from 1.0 A/g to 10 A/g within the voltage window of + 0.0 V to + 0.7 V are displayed in Fig. 8(a). The charging-discharging curve shape also confirmed that the CN electrode has a pseudocapacitive behavior. Fig. 8(b) displays the graph that plots the  $C_s$  value against the current density. The Eq. (2) assisted in calculating the  $C_s$  value of the CN electrode, which are 39.9 F/g, 36.5 F/g, 28.3 F/g, 23.6 F/g, 20.1 F/g and 17.3 F/g at current density of 1.0 A/g, 2.0 A/g, 4.0 A/g, 6.0 A/g, 8.0 A/g, and 10.0 A/g, respectively. Notably, the CN electrode exhibited capacitive behavior even at high current densities, demonstrating its suitability for energy storage applications. It is evident that the maximum  $C_s$  value of around 39.9 F/g was attained at a current density of 1.0 A/g. The  $C_s$  decrease as the current density rises because a high current density slows down the rate at which ions penetrate the electrode. Based on the GCD shown in Fig. 8(a), the energy and power density values of the CN electrode were calculated using Eqs. (3) and (4), respectively. Fig. 8(c) displays the Ragone graph of the energy and power density of the CN electrode. The highest energy and power density values of the CN electrode are 2.71 W.h/kg and 350 W/kg in 1.0 M KOH electrolyte. The electrode material based on the CN has a significant power density and an average energy density suggesting rapid charging and discharging



**Fig. 8.** (a) GCD curve at various current densities, (b) Specific capacitance variation with current density, (c) Ragone plot of CN electrode, (d) Cycling performance of CN electrode at a 1 A/g current density.

with minimal energy storage. Stability of electrode materials is a key consideration for prospective energy storage applications. Therefore, the CN electrode is evaluated using consecutive charge-discharge cycles with the results shown in Fig. 8(d). Electrochemical stability studies

were performed on the CN electrodes in 1.0 M KOH electrolytes at a current density of 1.0 A/g and showed excellent charge-discharge behavior and electrode durability with a capacitive retention of roughly 94 % after approximately 1000 cycles.



**Fig. 9.** (a) The digital picture of a green LED illuminated by the CN based symmetrical supercapacitor. (b) C-V curve of CN based symmetrical supercapacitor with a different voltage window at 200 mV/s scan rate, (c) GCD curve of CN based symmetrical supercapacitor at different voltage windows at a current density of 5 A/g, (d) C-V curve of CN based symmetrical supercapacitor at scan rates, (e) Specific capacitance variation with scan rates, (f) GCD curve of CN based symmetrical supercapacitor at different current density, (g) Specific capacitance variation with current density, (h) Ragone plot of CN based symmetrical supercapacitor, (i) Nyquist plot of CN based symmetrical supercapacitor from 1 mHz to 0.1 MHz before and after 1000 cycles, (j) Cycling performance of CN based symmetrical supercapacitor at a 2 A/g current density for 2000 cycles.

### 3.9. Electrochemical performance of CN-based symmetric supercapacitor device

The effectiveness of the CN-based symmetrical supercapacitor was studied by utilizing CV, GCD, and EIS, along with a two-electrode setup. Symmetrical supercapacitors, as utilized in LED (Fig. 9(a)), were fabricated with CN as electrode material, 1.0 M KOH electrolytes, and Whatman paper as a separator. Fig. 9(b) indicates the CV plot of the CN symmetric supercapacitor with the different potential ranges at a scan rate of 200 mV/s. It is worth mentioning here that the symmetric supercapacitor consistently displays an oxidation and reduction peak and exhibits pseudocapacitive behavior across all voltage windows. Ultimately, it was ascertained that the voltage range that allows the device to operate at its best was between  $-1.5$  and  $+2.2$  V. The symmetrical supercapacitor maintains outstanding electrochemical efficiency even when operating at a scan rate of 200 mV/s. More information about the properties of the CN symmetrical supercapacitor has been found by analyzing the GCD curves. The GCD curves measured under multiple potential windows, with a current density of 5 A/g, are displayed in Fig. 9(c). Fig. 9(d) shows a CV graph of CN-based symmetrical supercapacitor tests in a voltage window of  $-1.5$  V to  $+2.2$  V at multiple scan rates varying from 5 mV/s to 100 mVs. The CV plot of the CN symmetrical supercapacitor displays an oxidation and reduction peak, indicating pseudocapacitive behavior. The CV plot shows oxidation/reduction peak centered at  $+0.35$  V/ $-0.12$  V, respectively. The pseudocapacitive behaviour of CN symmetric supercapacitor is due to faradaic redox reactions caused by surface functional groups like nitrogen-containing groups. The conjugated  $\pi$ -electron system in g-C<sub>3</sub>N<sub>4</sub> allows for efficient electron transport and fast charge-discharge processes [28]. CN has a porous structure that provides maximal electrolyte exposure and serves as an effective base for electron and ion transport in supercapacitors. Hence, the CN symmetric supercapacitor may function at scan varying from 5 mV/s to 100 mV/s and different potential windows ranging from  $(-1.5$  V to  $+1.0$  V) to  $(-1.5$  V to  $+2.2$  V). The CN symmetric supercapacitor's current response was clearly increasing as scan rates increased, demonstrating the electrode's capacitive behavior in a way that was comparable with previously reported data [63]. Additionally, Eq. (5) was used to calculate the  $C_{cell}$  of a CN symmetric supercapacitor using a CV plot. Fig. 9(e) illustrates the relationship between  $C_{cell}$  and scan rate within the  $-1.5$  V to  $+2.2$  V potential window. When the CN symmetric supercapacitor was scanned at 5 mV/s, 10 mV/s, 25 mV/s, 50 mV/s, 75 mV/s and 100 mV/s, the  $C_{cell}$  values were 74.9 F/g, 64.9 F/g, 47.6 F/g, 37.6 F/g, 32.3 F/g, and 28.5 F/g, respectively. The  $C_{cell}$  decreased with increasing scan rate owing to diffusion limitations. At higher scan rates, electrolyte ions may not have enough time to permeate the electrode's active region, resulting in reduced charge storage. In contrast, decreasing the scan rate allows for deeper electrolyte penetration and increased charge storage [52].

The GCD curves at different current densities spanning from 0.5 A/g to 10.0 A/g recorded in the potential window between  $-1.5$  V to  $+2.2$  V are illustrated in Fig. 9(f). The form of the GCD curves indicates a rapid charging and discharging process with outstanding electrochemical capacitance characteristics, confirming the supercapacitor's pseudocapacitive behavior. The graph between the value of  $C_{cell}$  and current density is shown in Fig. 9(g). Eq. (6) is used to calculate the  $C_{cell}$  value of the symmetric supercapacitor device which are 149.1 F/g, 78.1 F/g, 44.5 F/g, 30.9 F/g, 26.5 F/g, 18.7 F/g, 14.4 F/g, and 11.6 F/g at a current density of 0.5 A/g, 1.0 A/g, 2.0 A/g, 3.0 A/g, 4.0 A/g, 6.0 A/g, 8.0 A/g and 10.0 A/g, respectively. It is significant to remark that capacitive behavior was detected even at high current densities, such as 10 A/g. It is evident that the maximum  $C_{cell}$  value of about 149.1 F/g was reached at a current density of 0.5 A/g. Fig. 9(h) displays the Ragone plot of the  $P_{cell}$  and  $E_{cell}$  of the CN-based symmetric supercapacitor in a two-electrode arrangement. The  $P_{cell}$  and  $E_{cell}$  values of the symmetric supercapacitor are estimated utilizing Eqs. (7) and (8), using

the GCD diagram and found to be 925 W/kg and 141.8 Wh/kg, respectively. The  $C_{cell}$  and  $E_{cell}$  value of the device is significantly higher than that of hollow mesoporous carbon spheres (130.5 F/g, 46.4 Wh/kg) [64], carbon xerogel (110 F/g, 4.5 Wh/kg) [65], carbon nitride supported ultra-fine bismuth sulfide (0.02435 Wh/kg) [66], B-rGO (120 F/g, 6.45 W h/ kg) [67], Co<sub>3</sub>O<sub>4</sub> (95 F/g, 29 Wh/kg) [68], Mo<sub>1.33</sub>CTz (140.4 F/cm<sup>3</sup>) [69] and pristine g-C<sub>3</sub>N<sub>4</sub> (34.8 F/g, 19.33 Wh/kg) [28] as shown in Table 1. L.G. Ghanem et al. [28] synthesized pristine g-C<sub>3</sub>N<sub>4</sub> using a simple, low-cost thermal polymerization method with post-synthesis treatment. Their g-C<sub>3</sub>N<sub>4</sub> exhibited a SSA of 32.51 m<sup>2</sup>/g and a total pore volume of 0.3 cm<sup>3</sup>/g. The resulting g-C<sub>3</sub>N<sub>4</sub>-based symmetric supercapacitor delivered a  $C_s$  of 34.8 F/g at a current density of 0.5 A/g in 0.5 M H<sub>2</sub>SO<sub>4</sub> electrolyte. In this study, the CN was synthesized using the same method but without post-treatment, aiming for a simpler and more cost-effective approach. This resulted in a higher SSA of 43.188 m<sup>2</sup>/g and a larger total pore volume of 2.64 cm<sup>3</sup>. Higher surface area offered more active sites for further electrochemical reactions, with improved performance. The resultant CN symmetric supercapacitor henceforth obtained higher  $C_{cell}$  values of 149.1 F/g at 0.5 A/g in the 1.0 M KOH electrolyte. Therefore, it is suggested that the CN synthesized without post-treatment in this study exhibited a higher SSA as compared to L.G. Ghanem's study [34]. The symmetric supercapacitor fabricated with CN without post-treatment exhibited remarkable power and energy density, enabling rapid charging and discharging while effectively storing energy. Further, EIS characteristics are studied to investigate the electrical conductivity and ion-transport characteristics of the CN-based symmetric supercapacitor. Fig. 9(i) displays the Nyquist plots for the symmetric supercapacitor device with an equivalent circuit. These plots were obtained in the 100 kHz to 0.1 Hz frequency span. The two-electrode symmetric supercapacitor is

**Table 1**

Comparison between the symmetric supercapacitor of CN and another symmetrical supercapacitor.

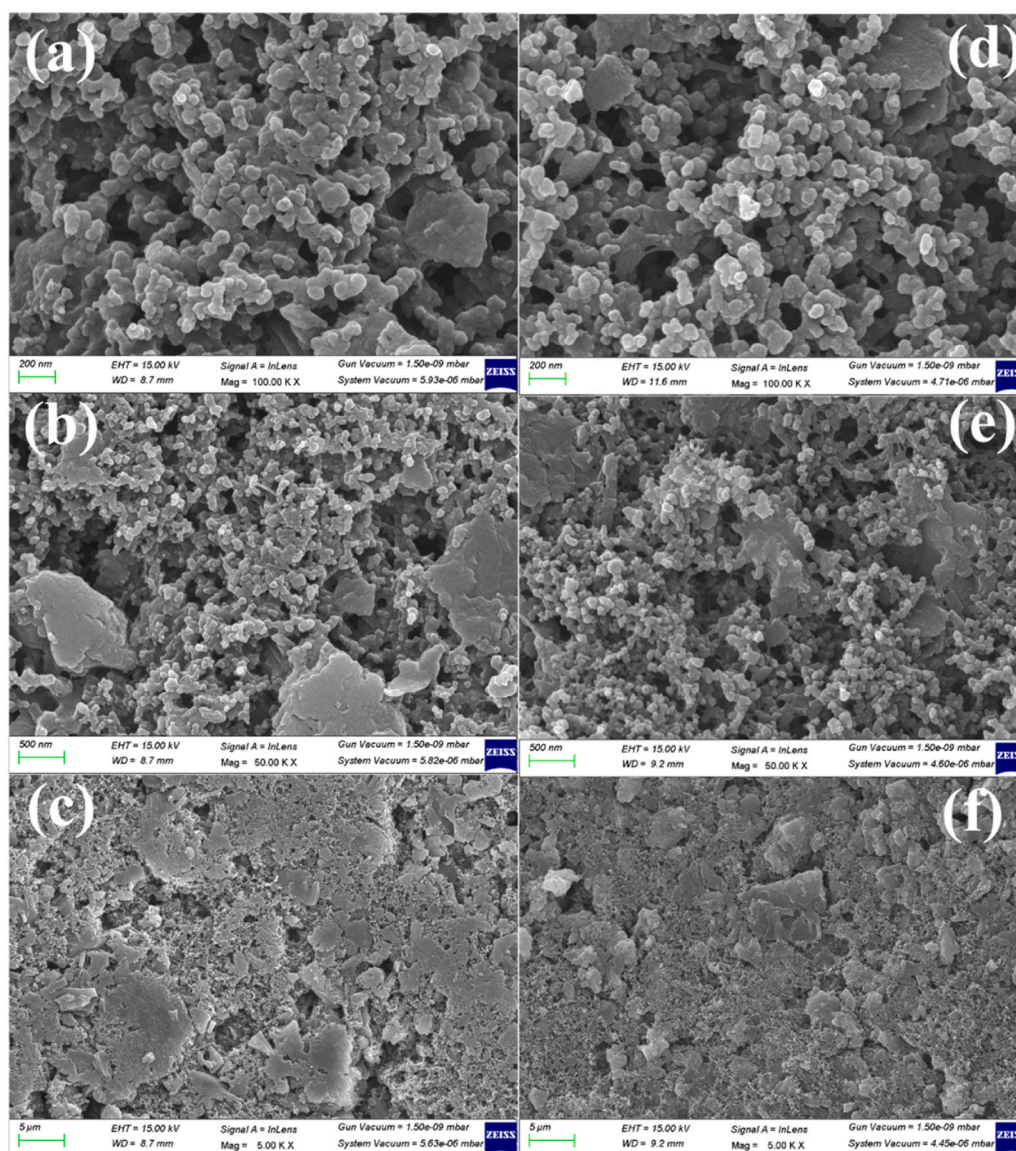
| Material                                            | Current density/ Scan rates | Specific capacitance    | Energy density                                       | Capacitance retention       | Ref.          |
|-----------------------------------------------------|-----------------------------|-------------------------|------------------------------------------------------|-----------------------------|---------------|
| Hollow Mesoporous Carbon Spheres                    | 0.2 A/g                     | 130.5 F/g               | 46.4 Wh/kg at 160 W/kg                               | 100 % after 2500 cycles     | [64]          |
| Carbon Xerogel                                      | 0.2 A/g                     | 110 F/g                 | 4.5 Wh/kg at 300 W/kg                                | 99 % after 10,000 cycles    | [65]          |
| B-rGO                                               | 0.1 A/g                     | 120 F/g                 | 6.45 W h/ kg at 8000 W/kg                            | 89.8 % after 2000 cycles.   | [67]          |
| Carbon Nitride Supported Ultra-fine Bismuth Sulfide | 0.02 A/g                    | -                       | 0.02435 Wh/kg at 186.57 W/kg                         | 99 % after 500 cycles       | [66]          |
| Co <sub>3</sub> O <sub>4</sub>                      | 6 mA                        | 112 F/g                 | 29 Wh/kg at 4745 W/kg                                | 91.40 % after 10,000 cycles | [68]          |
| Mo <sub>1.33</sub> CTz                              | 2 mV/s                      | 140.4 F/cm <sup>3</sup> | 38.2 mWh/cm <sup>3</sup> at 196.6 mW/cm <sup>3</sup> | 84.4 % after 20,000 cycles  | [69]          |
| Ti <sub>3</sub> C <sub>2</sub> Tz                   | 2 mV/s                      | ~92 F/cm <sup>3</sup>   | 41.3 mWh/cm <sup>3</sup> at 165.2 mW/cm <sup>3</sup> | -                           |               |
| Pristine g-C <sub>3</sub> N <sub>4</sub>            | 0.5 A/g                     | 34.8 F/g                | 19.33 Wh/kg at 499.9 W/kg                            | 126 % after 20000 cycles    | [28]          |
| CN                                                  | 0.5 A/g                     | 149.1 F/g               | 141.8 Wh/kg at 925 W/kg                              | 93.66 % after 1000 cycles   | In this study |

subjected to a + 5 mV OCV signal. The absence of a semi-circular curve at the higher frequencies indicated the exceptional electrical conductivity and low electrode internal and surface resistance in the symmetric supercapacitor. The Nyquist plot depicted a linear trend in the low-frequency domain that is not perpendicular to the Y-axis, which may result from the porous nature of the CN electrode. The porous structure of the CN electrode ensured complete electrolyte accessibility for ion adsorption and desorption [63]. The total resistance from the electrolyte, electrode, and electrical connections resulted in equivalent series resistance ( $R_{\text{ESR}}$ ), represented by the high-frequency area's intercept value. The symmetric supercapacitor device showed the  $R_{\text{ESR}}$  values of  $4.07 \Omega$  and the  $R_{\text{ct}} \sim 6.98 \Omega$ . The low values of  $R_{\text{ESR}}$  and  $R_{\text{ct}}$  indicate minimal internal contact resistance and speedy charge-discharge rate, aligning with the GCD findings [19]. The stability of symmetric supercapacitors is crucial for future energy storage applications. Therefore, a symmetric supercapacitor device was tested using 2000 consecutive charge-discharge using 1.0 M KOH electrolytes and applying a current density of 1.0 A/g as highlighted in Fig. 9(j). The cyclic performance of the device demonstrated exceptional charge-discharge behavior and excellent stability with a retention of

about 89.92 % after 2000 cycles. To demonstrate the utilization of the fabricated device in real-world situations, we successfully used our symmetrical supercapacitor device by connecting it to a green LED (2.2 V) and illuminating it, as shown in Fig. 9(a). CN-based symmetric supercapacitors showed unique electrochemical characteristics due to their microstructure, surface properties, and chemical composition mainly: (i) the porous nature of CN increased the surface area boosting the  $C_s$ , (ii) interconnected holes enabled the electrolyte penetration and ion diffusion facilitating charge transfer.

### 3.10. XPS and FE-SEM analysis of the symmetric supercapacitor device

To learn more about the stability of these electrode materials, the morphological and electronic structural analysis of the 2000 cycled charge-discharge CN symmetric supercapacitor electrode was carried out through XPS and FE-SEM techniques as additional research. According to an earlier study, the long-term stability of pseudocapacitive materials in supercapacitor applications may be limited [70–72]. Fig. 10 (a-f) shows the FE-SEM image of the electrode before and after the 2000<sup>th</sup> cycles with different magnifications. The FE-SEM analysis



**Fig. 10.** (a-c) The FE-SEM image of the CN symmetric supercapacitor before the stability test with different magnifications. (d-f) The FE-SEM image of the CN symmetric supercapacitor after the stability test with different magnifications.

revealed that the electrode was intrinsically porous and comprised of CN, with all chemical inclusions, including PVDF and activated carbon, uniformly distributed across the electrodes. These porous structures improve electrolyte diffusion while enhancing charge storage kinetics [73]. The FE-SEM images obtained before and after the 2000 charging-discharging cycles showed that the electrode surface underwent some minor surface modifications. However, neither the roughening and enlargement nor the fracture of the electrode material were particularly evident.

Furthermore, the XPS study of the electrode is carried which includes a comprehensive survey list of the elements present on the surface of these materials as shown in Fig. 11(a). The extensive survey showed the presence of potassium, oxygen, nitrogen, carbon, and fluorine in the electrode after 2000 cycles. The presence of potassium and fluorine in the symmetric supercapacitor is due to the use of KOH (electrolyte) and PVDF. Fig. 11(b) depicts the deconvolution of C1s spectra, with peaks at

284.82, 288.23, and 285.35 eV. The peak at 284.82 eV is most likely due to C-C/C-H bonds, which indicates the existence of  $sp^2$ -hybridized carbon atoms in the graphitic structure of CN and activated carbon [47]. The peak at 285.35 eV is most likely related to N-C=N bonds, indicating the existence of triazine units in the CN structure. The peak at 288.23 eV is possibly due to C=O bonding, which could result from carbon atom oxidation [48]. The deconvolution of the N 1s XPS spectrum exhibits peaks at 398.87 and 400.82 eV (Fig. 11(c)). The peak at 398.87 eV corresponds to  $sp^2$ -hybridized nitrogen atoms found in the triazine rings of graphitic carbon nitride. The peak at 400.82 eV is usually ascribed to tertiary nitrogen atoms (N-C<sub>3</sub>) or amino groups (NH<sub>2</sub>) in the CN framework [48,50]. Fig. 11(d) illustrates the deconvolution of the XPS K 2p spectra into two different peaks, 293.070 eV and 295.839 eV, respectively. The spin-orbit splitting of the K 2p core level is responsible for these peaks. The peaks at 293.070 eV and 295.839 eV are related to the K 2p<sup>3/2</sup> and K 2p<sup>1/2</sup> levels, respectively. A high-resolution XPS O1s

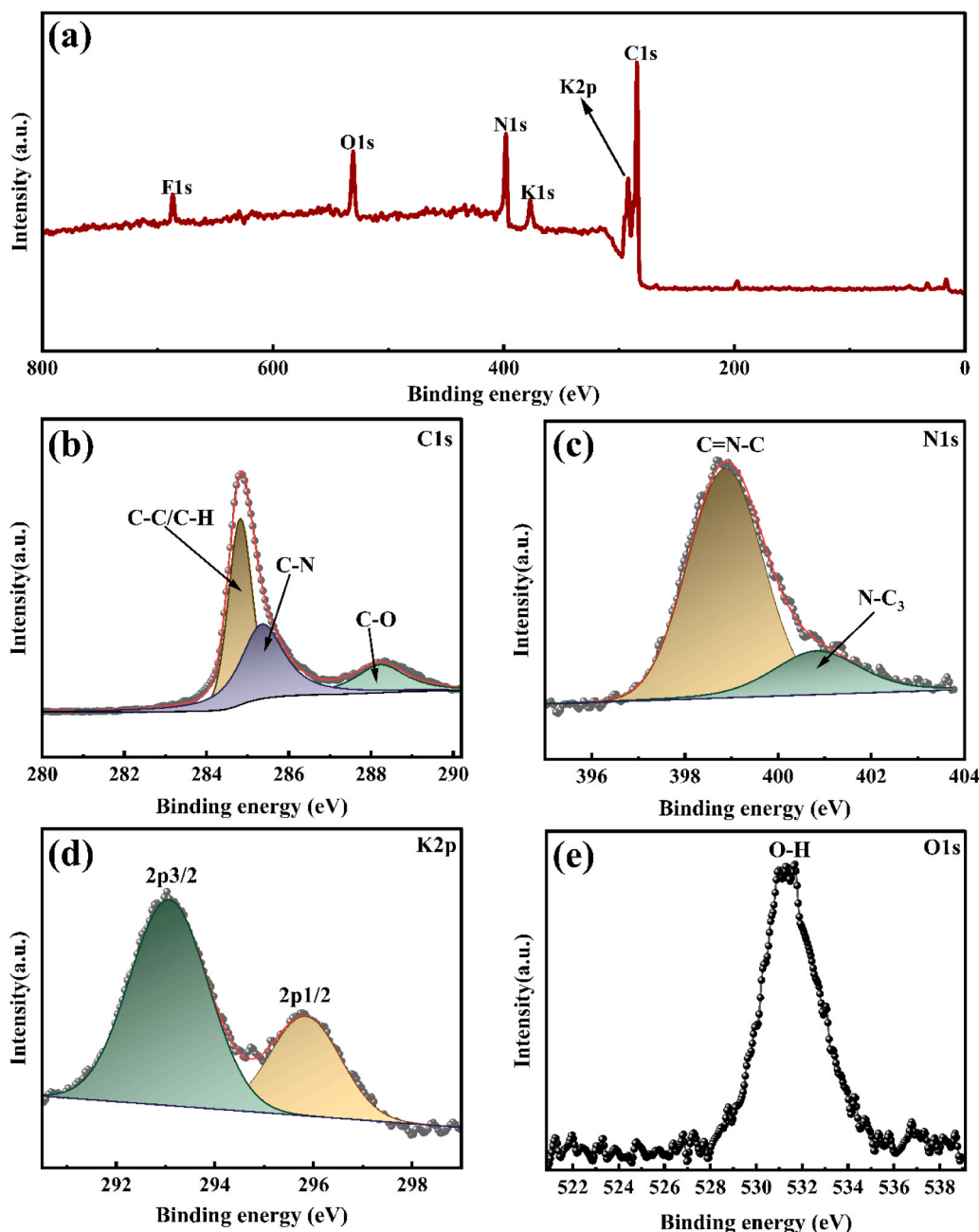


Fig. 11. (a) The XPS survey spectra of CN electrode after stability test, high-resolution spectra of (b) C1s, (c) N1s, (d) K2p, and (e) O1s.

spectrum is shown in Fig. 11(e), which indicates the existence of oxygen species that may be associated with surface oxidation and electrode interactions with the electrolyte. XPS investigations revealed that the composition of CN, comprising a chain of tri-s-triazine units, remained unchanged after the 2000 charging and discharging cycle. Thus, there is no conclusive proof of deterioration observed in the CN electrode, neither chemically nor morphologically. It was found consistent with the capacitive retention test, which demonstrated the stability of the CN symmetric electrode material.

#### 4. Conclusions

In brief, CN was prepared quickly and effectively using the thermal polycondensation method. The XRD shows the signature peaks of CN at  $13.10^\circ$  and  $27.30^\circ$ , confirming successful synthesis. The FTIR data demonstrate that CN has no extra functional group peak, indicating its purity. In a three-electrode arrangement, electrochemical studies were performed on the CN electrode. The outcomes showed the  $C_s$  to be around 35.2 F/g when using CV plot at a scan rate of 5 mV/s, and 39.9 F/g when using GCD plot at a current density of 1.0 A/g on the CN electrode. The CN electrode material demonstrated remarkable stability even after undergoing a thousand cycles, as evidenced by a  $C_s$  retention of more than about 94 %. Furthermore, symmetric supercapacitors based on CN were constructed, and the electrochemical properties of these supercapacitors were extensively investigated. Electrochemical tests of symmetric supercapacitors in two-electrode setups have shown that the  $C_{cell}$  of 149.1 F/g is achieved with GCD at a current density of 0.5 A/g. At a  $P_{cell}$  of 925 W/kg, the symmetric supercapacitor possessed an outstanding  $E_{cell}$  of 141.8 Wh/kg. The symmetric supercapacitor based on CN exhibited exceptional stability and charge-discharge behavior, retaining over 89.92 % of its capacitance even after 2000 cycles at a current density of 2.0 A/g. In summary, this study provides an in-depth examination of the structural and electrochemical characteristics of CN as well as the CN-based symmetric supercapacitor, providing valuable insights into the potential applications in energy storage systems. Further research on this study will aim to optimize the synthesis parameters and utilize other approaches to enhance CN surface area, porosity, conductivity etc., besides investigating whether functional groups or heteroatoms would improve the electrochemical performance. To further enhance the efficacy and attain synergetic integration, CN should be used in combination with other materials like conductive polymers, carbon nanotubes, and metal oxides.

#### CRediT authorship contribution statement

**Ranjeet Kumar Brajpuriya:** Formal analysis. **Shalendra Kumar:** Supervision, Formal analysis, Data curation, Conceptualization. **Kavita Kumari:** Formal analysis. **Bon-Heun Koo:** Formal analysis. **Saurabh Dalela:** Formal analysis. **P. A. Alvi:** Formal analysis. **Manas Nasit:** Formal analysis, Data curation, Conceptualization. **Ankush Vij:** Formal analysis.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Data availability

Data will be made available on request.

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