

Impact of separator on the electrochemical performance of g-C₃N₄/Co₃O₄ nanohybrid symmetric supercapacitors

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ABSTRACT

The present study aims to evaluate the influence of different separators on the electrochemical performance of a 2D g-C₃N₄ decorated Co₃O₄ symmetric supercapacitor. Exfoliated graphitic carbon nitride (ECN) was combined with Co₃O₄ to create nanocomposites in weight ratios of 1:0.01, 1:0.05, and 1:0.1 (COCN1, COCN2, COCN3). XRD analysis revealed that the ECN possesses a hexagonal structure, while Co₃O₄ exhibits a cubic spinel structure. COCN nanocomposites have been successfully synthesized, as confirmed by analyses using XRD and FTIR techniques. HR-TEM and SAED indicated that the Co₃O₄ adhered to the g-C₃N₄ matrix. The BET analysis revealed that the ECN and COCN2 show specific surface areas of 25 m²/g and 35 m²/g, respectively. The COCN2 nanocomposites attain a specific capacitance of 667.8 F/g at 1 A/g in 1.0 M KOH electrolyte, which is eight times more than ECN, due to the combined effects of the nitrogen content and cobalt oxidation states. The COCN2 exhibits an energy density of 45.45 Wh/kg at 218.75 W/kg power density, with 99.5 % capacitive retention. To explore the influence of different separators in symmetric devices, COCN2 was employed within the Swagelok assembly, featuring two separate types of separators: Whatman paper (COCN2/W/COCN2) and PVA-KOH gel electrolyte (COCN2/GE/COCN2). The COCN2/W/COCN2 device achieved a specific capacitance of 197.25 F/g at a current density of 1.0 A/g. Furthermore, it attained an energy density of 140.27 Wh/kg at a power density of 1600 W/kg, while retaining 94.23 % of its initial capacitance. In contrast, the COCN2/GE/COCN2 device showed a specific capacitance of 69.25 F/g and an energy density of 24.62 Wh/kg at 1600 W/kg, maintaining 95.4 % of its capacitance after 1000 cycles. The synergistic combination of Co₃O₄ and g-C₃N₄ offers a promising strategy for enhancing energy.

1. Introduction

In today's world of technological progress, the demand for sustainable and efficient energy storage solutions has significantly increased [1]. As a result, using renewable energy technology like solar cells, fuel cells, and wind turbines has become essential for producing electrical power. Nonetheless, the energy produced from renewable sources becomes inaccessible if not utilized in a timely manner. Consequently, it is essential to store this energy in suitable energy storage systems for future utilisation. At present, there is a strong focus on the advancement

of electrochemical energy storage systems aimed at balancing and adjusting energy supply while simultaneously reducing harmful emissions in the natural environment. Currently, a range of energy storage systems are in functioning, including batteries, fuel cells, and supercapacitors. Currently, supercapacitors are regarded as a leading option among the most promising energy storage technologies due to their remarkable power density (PD) and swift charge-discharge capabilities, and stand out as leading sustainable energy alternatives and strong contenders to fulfil this energy storage requirement [2]. Nonetheless, their energy density (ED) remains lower than that of conventional

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batteries, limiting their application in high-energy storage scenarios. To address this, researchers are investigating methods to boost the ED of supercapacitors. A promising strategy involves developing novel electrode materials with enhanced electrochemical properties [3]. So far that researcher are working on the two-dimensional (2D) materials, including MoS_2 , graphene, covalent organic frameworks (COFs), MXenes, metal-organic frameworks (MOFs), and graphitic carbon nitride ($\text{g-C}_3\text{N}_4$), have gained attention due to their superior conductivity, large surface area, and capacity to enhance ion diffusion, addressing pure metal oxides/hydroxides limitations [4–8].

Among them, $\text{g-C}_3\text{N}_4$ is significant due to its cost-effectiveness, straightforward synthesis, and the large number of pyridine-like nitrogen active sites that are crucial in redox reactions, and the π -conjugated covalent bonds between carbon and nitrogen improve the conductivity of $\text{g-C}_3\text{N}_4$. The hexagonal lattice structure of tri-s-triazine units in $\text{g-C}_3\text{N}_4$ provides numerous active sites for electrochemical activities [9], and the nitrogen-containing functional groups enhance its interaction with electrolyte ions, facilitating efficient charge transfer. Many researchers have shown that $\text{g-C}_3\text{N}_4$ can be synthesized through cost-effective thermal polymerization for supercapacitor applications. In our previous studies, the $\text{g-C}_3\text{N}_4$ was synthesized via thermal polymerization of urea, had a specific capacitance (C_{sp}) of 39.9 F/g at 1.0 A/g current density (CD) through galvanostatic charge-discharge (GCD) analysis in a three-electrode setup. The $\text{g-C}_3\text{N}_4$ -based symmetric supercapacitor device (SSCD) exhibited C_{sp} of 149.1 F/g at 0.5 A/g in two electrode configurations [10]. Devi et al. utilized autoclave-assisted solvent-free synthesis to create g-CN , achieving C_{sp} of 138.3 F/g at CD of 2.0 A/g, with 100 % retention over 5000 cycles, demonstrating its chemical stability and good electrochemical performance [11]. Wang et al. developed 3D network porous $\text{g-C}_3\text{N}_4$ layers achieving C_{sp} of 219.4 F/g at 20 A/g through simple calcination. The high capacitance stems from its extensive surface area, nitrogen doping, significant graphitic content, and hierarchical porous architecture [12]. Raghupati et al. used condensation to produce exfoliated $\text{g-C}_3\text{N}_4$ nanoflakes, further exfoliated using ultrasonication. They observed C_{sp} of 160 F/g at 1 A/g, retaining 96.2 % capacity after 3000 cycles, and reported that nano-flake shape enhances charge diffusion kinetics, improving storage mechanism [13]. These results indicated that exfoliated $\text{g-C}_3\text{N}_4$ has better electrochemical properties than bulk $\text{g-C}_3\text{N}_4$. While $\text{g-C}_3\text{N}_4$ has many advantages, it has several disadvantages, such as low electrical conductivity, aggregation, and fewer active sites than metal oxides, which limit its use as a supercapacitor.

To overcome these disadvantages, the investigation of $\text{g-C}_3\text{N}_4$ /nanocomposites was conducted. As it already discussed that metal oxides have good active sites for redox reactions, unique morphological traits, and high theoretical C_{sp} [14]. Various metal oxides, including Co_3O_4 , NiO , MnO_2 , Mn_2O_3 , and SnO_2 , have been studied for their robust electrochemical properties and high C_{sp} . [15]. There are numerous studies on cobalt-based materials to improve their electrochemical properties [16–19]. So, Researchers are particularly intrigued by Co_3O_4 because of its spinel structure and outstanding magnetic and electronic properties. As a result, it has a wide range of applications, such as lithium-ion batteries, supercapacitors, gas sensors, and catalytic materials. It is low-cost, electroactive, and has a high theoretical C_{sp} (3600 F/g). Furthermore, Co_3O_4 is interesting because of its unique structural features and higher theoretical capacitance compared to other metal oxides, making it an ideal candidate for supercapacitor electrode materials because of the presence of multiple oxidation states, namely Co^{2+} and Co^{3+} . These oxidation states facilitate faradaic redox reactions at the interfaces between the electrode and the electrolyte ions [20]. Co_3O_4 can be engineered into various structures, such as nanoparticles, nanowires, and nanoflowers via techniques like hydrothermal synthesis, microwave processing, solvothermal methods, and co-precipitation, to boost its electrochemical performance [21]. Kalpana et al. utilized hydrothermal synthesis to create a flower-like structure of Co_3O_4 particles, while spherical Co_3O_4 particles were formed through co-precipitation

synthesis. The findings showed that the spherical Co_3O_4 exhibited a C_{sp} of 366 F/g, whereas the flower-like Co_3O_4 displayed 233 F/g at a CD of 0.5 A/g [22]. employed the co-precipitation technique to synthesize elliptical Co_3O_4 , which exhibited a C_{sp} of 481 F/g at a CD of 1.0 A/g [23]. In contrast, Babu et al. utilized a hydrothermal synthesis approach to create an SSCD composed of cubic Co_3O_4 nanostructures, achieving an ED of 77.3 Wh/kg with a PD of 1997.7 Wh/kg [24].

Integrating $\text{g-C}_3\text{N}_4$ with Co_3O_4 in nanocomposites can create synergistic effects that significantly enhance the electrochemical performance of hybrid electrode materials. The interaction between these two components improves electrical conductivity, increases the active surface area, facilitates charge transfer, and enhances cycle stability [25]. The use of nitrogen-rich $\text{g-C}_3\text{N}_4$ alongside Co_3O_4 is expected to boost the electrochemical properties of the Co_3O_4 sample [26]. By reducing the tendency of Co_3O_4 nanoparticles to aggregate and by preventing structural degradation, the combination of $\text{g-C}_3\text{N}_4$ and Co_3O_4 can improve the cycle stability of the electrode material [27]. Thus, $\text{g-C}_3\text{N}_4/\text{Co}_3\text{O}_4$ nanocomposites present an effective method to enhance the electrochemical capacity of supercapacitors. Various research groups have actively explored the electrochemical properties of $\text{g-C}_3\text{N}_4/\text{Co}_3\text{O}_4$ nanocomposites. S. Yan and colleagues synthesized a hybrid of Co_3O_4 with $\text{g-C}_3\text{N}_4$ material. The composite electrode material exhibits superior electrochemical performance when compared to pristine Co_3O_4 electrodes [28]. As a result, the composite material demonstrates an extended discharge duration and enhanced capacity, thus improving overall capacitance performance. Following prolonged cycling, the composite material exhibits a remarkable capacity retention rate of 90.4 %. Yewale and his team developed a Co_3O_4 - $\text{g-C}_3\text{N}_4$ composite electrode that showed a C_{sp} of 198 F/g. The asymmetric supercapacitor made from this composite had a C_{sp} of 43 F/g and an ED of 11.86 Wh/kg [27]. Y. Zhang's team synthesized the $\text{Co}_3\text{O}_4/\text{g-C}_3\text{N}_4$ composite using a two-step process that included in-situ sedimentation and a carbon-assisted method. Electrochemical evaluations indicated that the $\text{Co}_3\text{O}_4/\text{g-C}_3\text{N}_4$ composite reached a C_{sp} of 1071 F/g at a CD of 1 A/g and, an asymmetric supercapacitor constructed from $\text{Co}_3\text{O}_4/\text{g-C}_3\text{N}_4//\text{AC}$ achieved a maximum ED of 20.4 Wh/kg [26]. Subbiah and his team created a high-performance supercapacitor by combining mesoporous Co_3O_4 with $\text{g-C}_3\text{N}_4$ nanosheets through a hydrothermal method, achieving a C_{sp} of 455 F/g, and SSCD showed an ED of 28.13 Wh/kg and a PD of 1.68 kW/kg [25].

In this study, $\text{g-C}_3\text{N}_4$ was produced through thermal polymerization, followed by ultrasonication to exfoliate the bulk $\text{g-C}_3\text{N}_4$, resulting is referred to as ECN. Co_3O_4 was prepared using the co-precipitation method, and ECN/ Co_3O_4 nanocomposites were formed through the ultrasonication technique. The structural features of these nanocomposites were characterized using XRD. Field emission scanning electron microscopy (FESEM) was employed to investigate the size distribution and morphology of the nanocomposites. X-ray photoelectron spectroscopy (XPS) provided insights into the electronic structural properties. The electrochemical performance of the nanocomposites was assessed using cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), and galvanostatic charge-discharge (GCD). The optimal nanocomposite sample was chosen to construct a SSCD using a Swagelok assembly. Two separators were used to develop SSCD: one utilized Whatman paper as the separator, while the other used a gel electrolyte as the separator. The novelty of this research is to evaluate the role of the separator on the electrochemical performance of the ECN/ Co_3O_4 nanocomposites SSCD. The performance of both SSCD was evaluated using CV, GCD, and EIS.

2. Experimental section

2.1. Synthesis of $\text{g-C}_3\text{N}_4$

The bulk synthesis of $\text{g-C}_3\text{N}_4$ was achieved through the thermal polymerization technique [29]. A quantity of 10g of raw melamine was

placed in an alumina crucible and subjected to heating at 500 °C for 3 h in a muffle furnace. The resulting fluffy powder was then combined with de-ionized (DI) water, and this mixture underwent sonication for 2 h to exfoliate the bulk g-C₃N₄ [13]. Subsequently, the solution was filtered, and the remaining material was dried in a hot-air oven to obtain a powder. The exfoliated g-C₃N₄, referred to as ECN, was then utilized for the synthesis of nanocomposites.

2.2. Synthesis of Co₃O₄

Cobalt oxide nanoparticles were produced using the co-precipitation technique [22]. A 0.5 M solution of cobalt nitrate was made with deionized water. Subsequently, a 0.2 M NaOH solution was gradually added until the solution's pH reached 7. This mixture was then stirred continuously overnight. The precipitate formed was washed and centrifuged with deionized water and ethanol. The resulting precipitate was dried overnight and then calcined at 550 °C for 3 h. The final product, Co₃O₄, was ground into a fine powder for subsequent nanocomposite synthesis.

2.3. Synthesis of g-C₃N₄/Co₃O₄

The g-C₃N₄/Co₃O₄ nanocomposites were produced using a simple ultrasonication method [29,30]. Initially, separate dispersions of ECN and Co₃O₄ were prepared in DI water at different weight ratios. Each dispersion was stirred for an hour before being mixed together, stirred again, and sonicated for an additional 2 h. After this process, the mixture was washed, centrifuged, and dried to obtain the g-C₃N₄/Co₃O₄ nanocomposite. These nanocomposites are designated as COCN1, COCN2, and COCN3, corresponding to ECN: Co₃O₄ weight ratios of 1: 0.01, 1: 0.05, and 1: 0.1, respectively. Fig. 1 depicts the synthesis procedure.

2.4. Instrumentation and characterization

The examination of the crystal structure and phase purity of g-C₃N₄/Co₃O₄ nanocomposites were performed using the Bruker D8 Advance EcoPro diffractometer, which utilizes Cu K α radiation ($\lambda = 1.54056 \text{ \AA}$). The identification of functional groups within the samples was achieved using Fourier transform infrared (FT-IR) spectroscopy, which employed a PerkinElmer FT-IR spectrometer. A comprehensive analysis of the nanocomposites morphology and microstructure was carried out using a high-resolution FE-SEM, specifically the JEOL JSM-7900F model. The surface chemical composition was evaluated through XPS, conducted with a Kratos Analytical AXIS SUPRA + instrument. The XPS analysis was performed using a monochromatic Al K α X-ray source at 225 Watts, with a step size of 1.0 eV and a sweep time of 120 s.

2.5. Electrochemical measurement

The electrochemical characteristics of the ECN, Co₃O₄, COCN1, COCN2, and COCN3 electrodes were assessed using a Biologic VSP-300. The electrochemical tests were conducted with a standard three-electrode system, comprising an Ag/AgCl reference electrode, a platinum wire counter electrode, and the working electrode being studied. To prepare the working electrode, the synthesized material, carbon black (as a conductive agent), and PVDF (as a binder) were combined in a ratio of 80:10:10. This mixture was then blended with NMP to form a slurry. The slurry was applied to a 5 mm $\times$ 5 mm Ni foam substrate through drop-casting and subsequently dried for 12 h. Electrochemical evaluations, such as C_{sp}, ED, PD, and cyclic stability, were conducted using CV, GCD, and EIS methods in a 1.0 M electrolyte solution.

2.6. Gel electrolyte synthesis

To synthesize the gel electrolyte, 20 g of PVA and 10 g of KOH were dissolved in 300 ml of DI water [31,32]. The solution was continuously stirred at 90 °C for 8 h to ensure the PVA was fully dissolved. After achieving a homogeneous solution, it was poured into a Petri dish, with the volume adjusted to obtain the desired thickness for the separator. The Petri dish was then left to dry at room temperature overnight, allowing the gel electrolyte to solidify. Fig. 2 illustrates the complete synthesis process of the gel electrolyte.

3. Results and discussion

3.1. XRD analysis

The phase identification and structural characteristics of ECN, Co₃O₄, and COCN nanocomposites were examined using XRD patterns. The Co₃O₄ diffraction peaks appear at 2θ angles of 19.04°, 31.34°, 36.93°, 44.89°, 59.47°, and 65.36°, corresponding to the (111), (220), (311), (400), (511), and (440) planes, respectively [22]. Further, the FullProf program facilitated Rietveld refinement was used to analyze the XRD pattern of pure Co₃O₄ nanoparticles. A pseudo-voigt function and CIF file of Co₃O₄ (COD ID: 9005896) was employed to fit the data, showing a strong correlation between the calculated and observed intensities. Fig. 3(a) presents the Rietveld refinement of Co₃O₄, confirming the pure cubic phase with lattice parameters $a = b = c = 8.08 \text{ \AA}$, phase angles $\alpha = \beta = \gamma = 90^\circ$, and a space group of Fd-3m [28]. The refinement produced a χ^2 value of 1.07, indicating a highly precise fit. The crystal structure obtained from the refinement data, visualized with VESTA software, is depicted in Fig. 3(b). Fig. 3(c) displays the XRD patterns of ECN, Co₃O₄, and the COCN nanocomposite. Co₃O₄ shows a face-centred

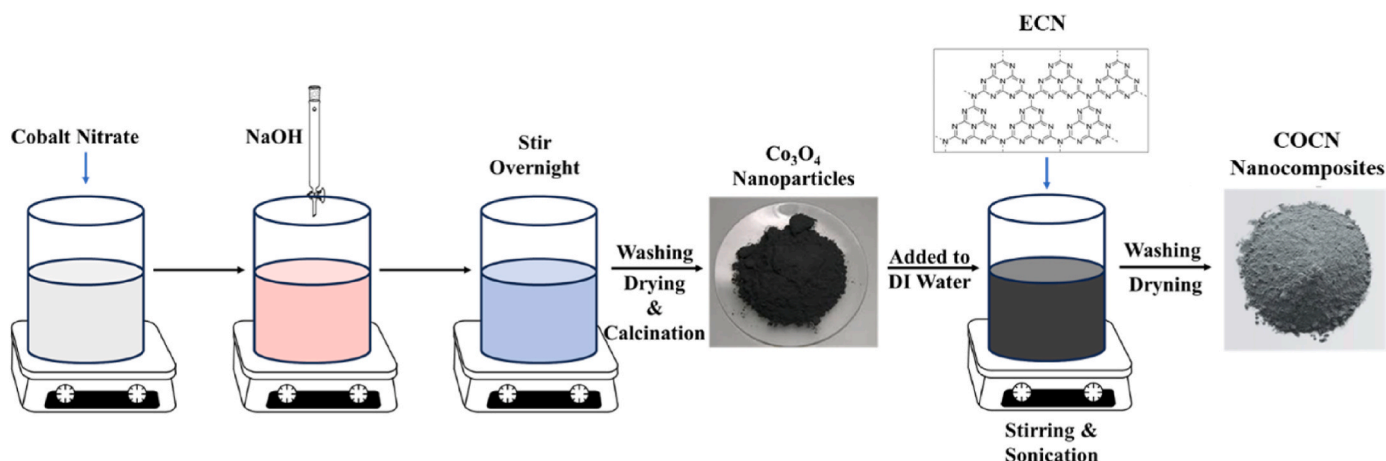


Fig. 1. Synthesis of Co₃O₄ and COCN nanocomposites.

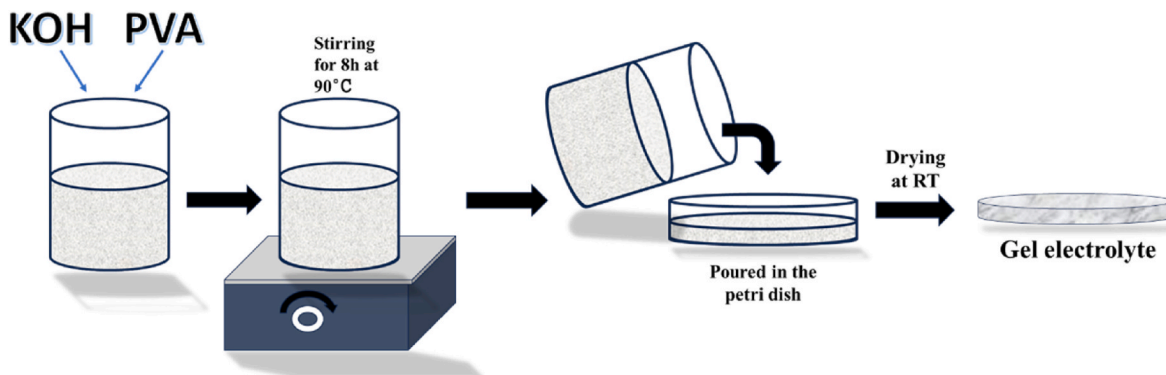
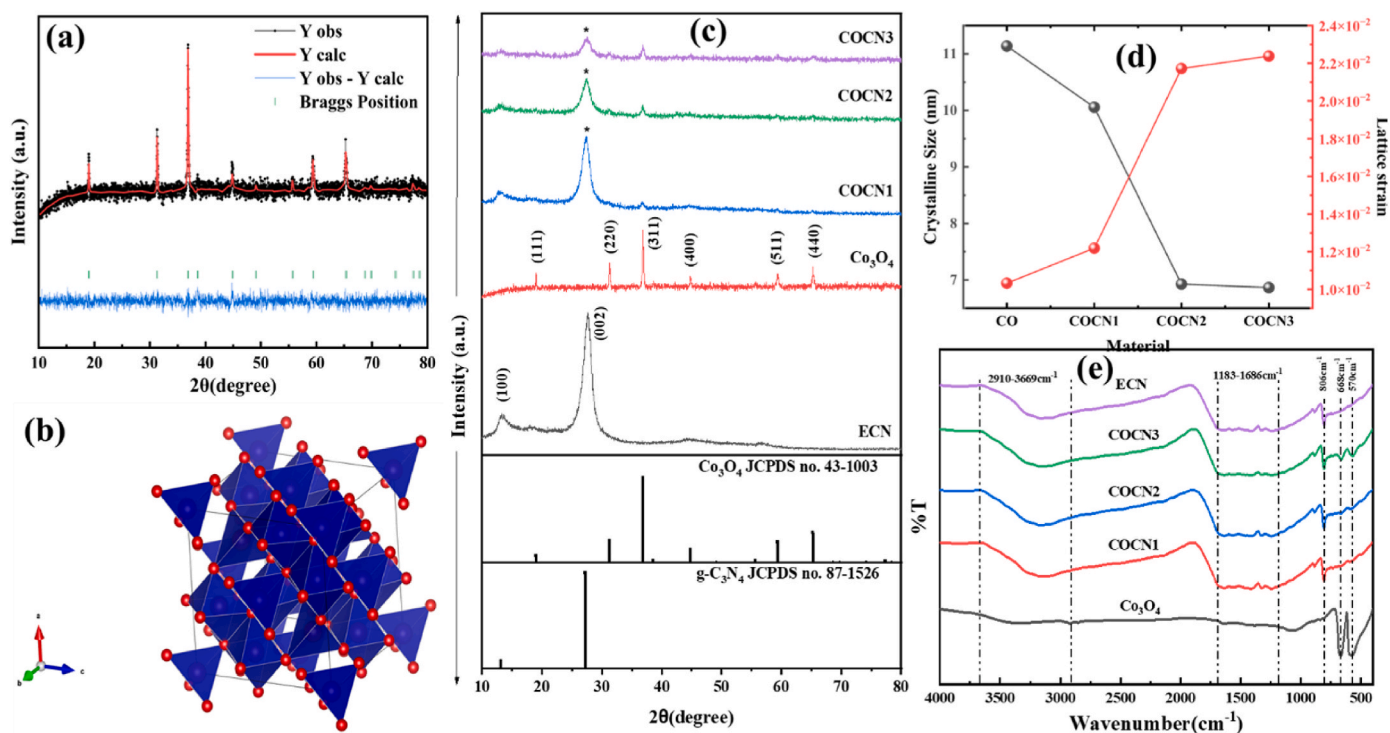


Fig. 2. Synthesis of gel electrolyte using PVA and KOH.

Fig. 3. (a) Rietveld refinement of Co_3O_4 , (b) Crystal structure of Co_3O_4 using VESTA, (c) XRD pattern of ECN, Co_3O_4 , COCN1, COCN2, and COCN3, (d) Comparison of crystalline size and lattice strain, (e) FTIR pattern of ECN, Co_3O_4 , COCN1, COCN2, and COCN3.

spinel cubic phase, while ECN exhibits a hexagonal phase [22]. The XRD patterns of Co_3O_4 and ECN align with JCPDS card numbers 87–1526 and 43–1003, respectively. The absence of additional peaks in the XRD pattern confirms the single-phase nature of the synthesized materials. For ECN, the prominent diffraction peak at $2\theta = 27.66^\circ$ corresponding to the (002) plane indicates interlayer stacking of the C–N framework within its graphitic structure [33]. The COCN nanocomposite shows diffraction peaks of $\text{g-C}_3\text{N}_4$ and Co_3O_4 , indicating the successful synthesis of COCN nanocomposites [26]. The crystallite size was calculated using the dominant (311) plane peak in the XRD pattern, applying the Debye-Scherrer formula (Eq. (1)) [34]:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

here, D represents the crystallite size, K denotes the shape factor, λ is the wavelength of the X-ray source ($1.5406 \text{ \AA}$), β stands for the full width at half maximum (FWHM) of the (311) peak in radians, and θ is the Bragg diffraction angle. The crystallite sizes calculated for Co_3O_4 , COCN1,

COCN2, and COCN3 were 10 nm, 9 nm, 6.9 nm, and 6.8 nm, respectively. A key element in the structural enhancement of the nanocomposites is the improved distribution and interaction of Co_3O_4 within the ECN, as evidenced by the decrease in crystallite size. As the crystallite size decreases, the surface area increases, which is directly linked to the material's electrochemical properties. Consequently, a larger surface area provides more active sites, thereby enhancing electrochemical performance. Therefore, materials with smaller sizes typically demonstrate superior electrochemical properties. The lattice strain of the material was calculated using Eq. (2), which is based on the Williamson-Hall (W–H) method [35]:

$$\beta \cos \theta = \frac{K\lambda}{D} + 4\epsilon \sin \theta \quad (2)$$

The symbol ϵ denotes the lattice strain. The calculated lattice strain values for Co_3O_4 , COCN1, COCN2, and COCN3 are 1.03×10^{-2} , 1.21×10^{-2} , 2.17×10^{-2} , and 2.23×10^{-2} , respectively. Fig. 3(d) shows a comparison between crystalline size and lattice strain. An increase in lattice strain with higher Co_3O_4 concentration suggests more significant

lattice distortion due to the strong interaction between Co_3O_4 and ECN. This distortion is likely to improve the structural and electrochemical properties of the nanocomposites by increasing active sites and enhancing charge transfer. The combined effects of smaller crystallite size and increased lattice strain imply that COCN nanocomposites, especially COCN2 and COCN3, have optimized structural characteristics. These characteristics suggest that COCN nanocomposites could be a promising option for energy storage applications.

3.2. FTIR analysis

Fig. 3(e) illustrates the FTIR spectra for ECN, Co_3O_4 , and the COCN nanocomposite. In the FTIR spectra of Co_3O_4 , the vibration bands detected at 668 cm^{-1} and 570 cm^{-1} are ascribed to the stretching vibrations of the O–Co–O bonds. Furthermore, the peak at 565 cm^{-1} is associated with the stretching vibration of Co–O [36]. In the ECN spectra, a notable peak at 810 cm^{-1} indicates the bending vibration of the heptazine unit. The broadband observed at $2910\text{--}3669\text{ cm}^{-1}$ is linked to the stretching vibration of the –OH bond [25]. Furthermore, several peaks ranging from 1183 cm^{-1} to 1686 cm^{-1} correspond to the stretching vibrations of aromatic C–N and C=N units. The nanocomposites display characteristic peaks from both materials, indicating the successful integration of Co_3O_4 into ECN sheets. The amount of Co_3O_4 present in the COCN nanocomposite influences the intensity of these vibrational bands [37]. Furthermore, the unchanged peak at 810 cm^{-1} suggests that the solid framework of the tri-s-triazine ring remains intact. These findings indicate that Co_3O_4 can be successfully anchored onto the ECN exfoliated sheet through ultrasonication without significantly disrupting the original structure of the ECN.

3.3. SEM analysis

Fig. 4(a, b, c) displays FESEM images of ECN, Co_3O_4 , and COCN2. In Fig. 4(a), the FESEM analysis of ECN reveals a sheet-like exfoliated

morphology with a rough and porous structure. The exfoliation process leads to a more porous sheet structure in ECN, which enhances electron transport during electrochemical processes, thereby optimizing the electrochemical properties and improving the supercapacitor's performance [38]. Fig. 4(b) shows the FESEM images of Co_3O_4 , indicating that the material synthesised via co-precipitation has an irregular elliptical shape. The smaller size of Co_3O_4 nanoparticles increases the active surface area and electrochemical properties of the material, which will be further explored in the electrochemical study [39]. Fig. 4(c) presents the FESEM image of COCN2, highlighting a porous structure and a rough surface. As previously mentioned, this porous structure and rough surface allow for easy penetration of electrolyte ions, potentially enhancing its electrochemical properties. Additionally, Fig. 4(d–f–i) shows the element mapping images of COCN2. These images depict the presence of various components in the sample: Fig. 4(d) illustrates a combination of C (lemon color), N (orange color), Co (blue color), and O (red color); Fig. 4(f–i) provides individual images of C in lemon, N in orange, O in red, and Co in blue, respectively. Fig. 4(e) displays the EDS spectrum of COCN2, confirming the presence of C, N, Co, and O, thus verifying the formation of COCN2.

3.4. TEM analysis

It is worth mentioning here that the COCN2 nanocomposite exhibited the highest C_{sp} among all the samples, as revealed by the electrochemical studies. Consequently, HR-TEM and SAED analyses were performed to gain a deeper insight into the microstructure and phase characteristics of the COCN2 nanocomposite. Fig. 5(a) depicts the HR-TEM images of the COCN2 nanocomposite, highlighting the presence of Co_3O_4 nanoparticles with an elliptical shape that are consistently distributed throughout the ECN, corroborating the SEM observations. HRTEM micrographs were employed to develop the size distribution histogram, and ImageJ software was utilized to assess the particle size. The particles display a broad range of sizes, with an average diameter of

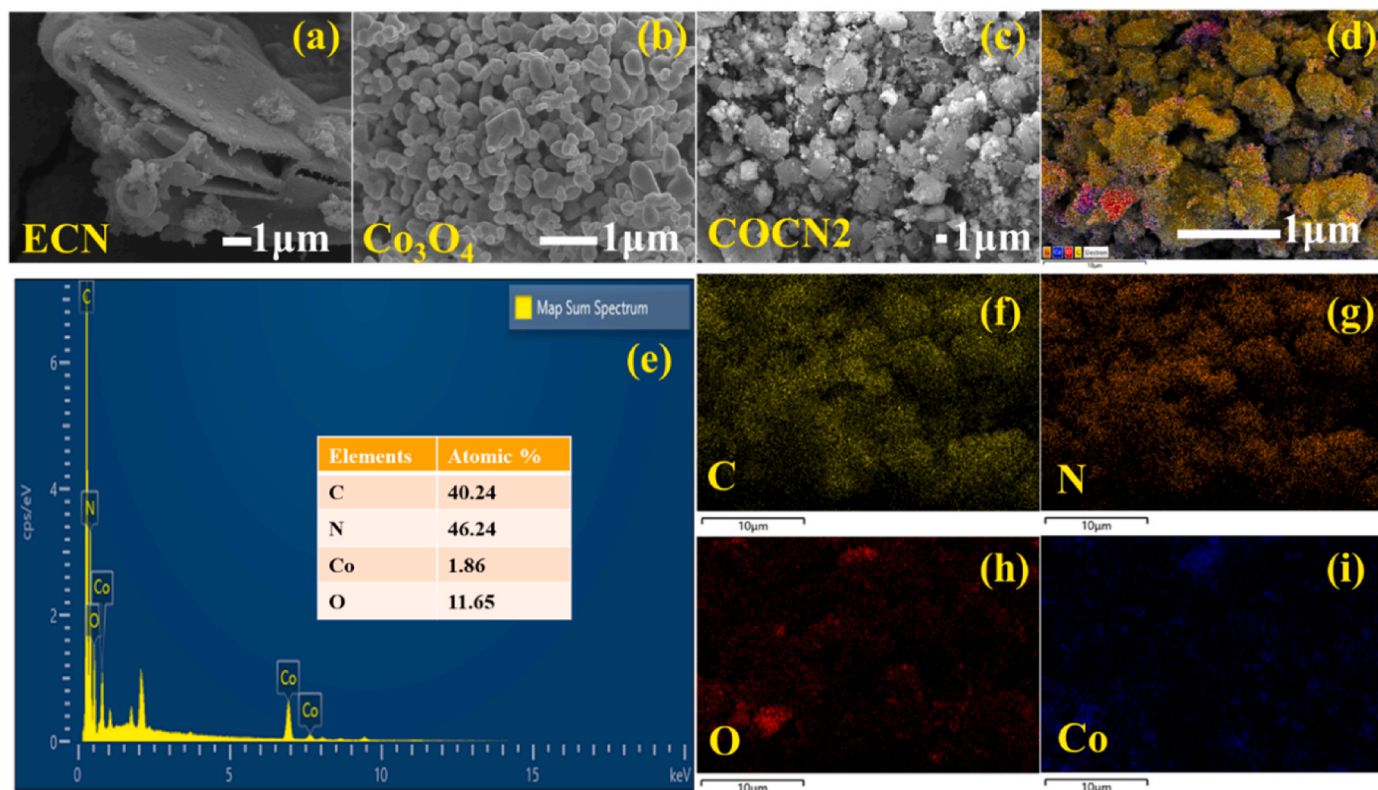


Fig. 4. FESEM images of (a) ECN, (b) Co_3O_4 , (c) COCN2, (e) EDS spectrum of COCN2, (f–i) element mapping of COCN2.

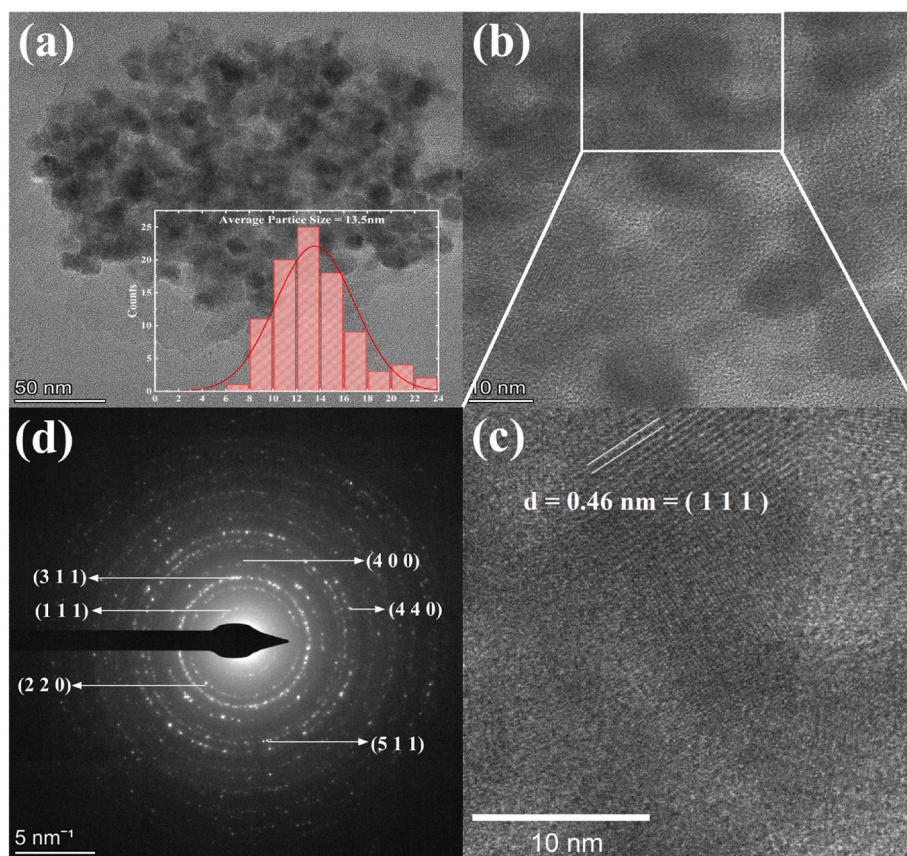


Fig. 5. (a–b) HR-TEM micrograph of COCN2, (c) FFT image of COCN2, (d) SAED pattern of COCN2.

about 13.5 nm. Moreover, Fig. 5(b) illustrates the HR-TEM micrographs of COCN2, while Fig. 5(c) provides an enlarged depiction of this image, which was instrumental in calculating the interplanar distance (d). The HR-TEM micrograph reveals that the (111) plane of Co_3O_4 in the COCN2 nanocomposite exhibits a d-spacing of 0.46 nm. Fig. 5(d) displays the SAED pattern of the COCN2 nanocomposite, featuring six distinct diffraction rings corresponding to the (111), (220), (311), (400), (511), and (440) planes. These reflections confirm the presence of Fd-3m spinel cubic symmetry in the Co_3O_4 phase, thereby verifying the crystalline structure and structural integrity of the nanocomposite [40].

3.5. XPS analysis

A thorough investigation into the electronic structure of the COCN2 composite was carried out using XPS, attributed to its outstanding C_{sp} . Fig. 6(a) displays the survey scan spectra for the COCN2 nanocomposite. The spectrum reveals distinct peaks that correspond to cobalt, oxygen, carbon, and nitrogen, with their respective binding energies recorded at 77.27 eV, 528.27 eV, 284.27 eV, and 398.27 eV. This confirms the presence of all these elements within the COCN2 nanocomposite. Fig. 6(b) illustrates the deconvoluted N 1s spectra, which display peaks at 394.96 eV, 395.61 eV, and 397.36 eV. These peaks correspond to the C–N=C, N–C₃, and C–N–H bonds, respectively [26,27,41]. These findings underscore the significant role of nitrogen in the ECN nanosheet, as it is essential for improving conductivity and promoting charge transfer within the nanocomposite. Fig. 6(c) presents the deconvoluted carbon spectra, revealing three distinct peaks at 281.37 eV, 282.71 eV, and 284.61 eV, which are associated with C–C, C=O, and N–C=N bonds, respectively [25,27]. These peaks demonstrate the diverse carbon bonding scenarios within the COCN2 nanocomposite. The presence of C=O and N–C=N bonds suggests interactions between carbon and oxygen, as well as nitrogen, within the ECN structure, while the C–C bonds

signify the basic carbon framework. Fig. 6(d) highlights the Co 2p spectra, showcasing both the convolution and deconvolution peaks. The Co 2p_{3/2} and Co 2p_{1/2} states are represented by two primary peaks at 778.98 eV and 794.07 eV, respectively, illustrating the spin-orbit splitting typical of cobalt in Co_3O_4 nanoparticles. Peaks at 776.85 eV and 791.81 eV verify the presence of the Co^{3+} state, while additional peaks at 778.46 eV and 793.61 eV suggest the Co^{2+} state [42]. The simultaneous presence of Co^{2+} and Co^{3+} oxidation states in the nanocomposite indicates a mixed-valence system, which is essential for redox reactions [43]. These features suggest an improvement in the pseudocapacitive performance of the COCN2 nanocomposite [41]. Fig. 6(e) depicts the deconvoluted oxygen spectra, revealing three prominent peaks at 526.90 eV, 528.46 eV, and 530.06 eV. The peak at 526.90 eV signifies the presence of the C–N–O bond, while the one at 528.46 eV is associated with Co–O bonds [25]. The peak at 531.21 eV clearly indicates the presence of oxygen ions and adsorbed water molecules within the nanocomposite, which could potentially enhance ionic conductivity and improve the electrochemical performance of COCN2 nanocomposites. The detailed XPS analysis provides in-depth insights into the chemical states and bonding environments of carbon, nitrogen, cobalt, and oxygen in the COCN2 nanocomposite.

3.6. BET analysis

Fig. 7 presents the adsorption and desorption isotherms for the COCN2 and ECN samples across a range of pressures (p/p_0 , 0–1). The adsorption and desorption profiles for the ECN and COCN2 materials depicted in Fig. 7(a) are indicative of Type III adsorption isotherms. Fig. 7(b) showcases the nanocomposite of ECN and COCN2. Type III isotherms are marked by low surface energy and weak interactions between the adsorbate molecules and the surface [44]. This trait implies that these materials can develop multiple monomolecular layers, with

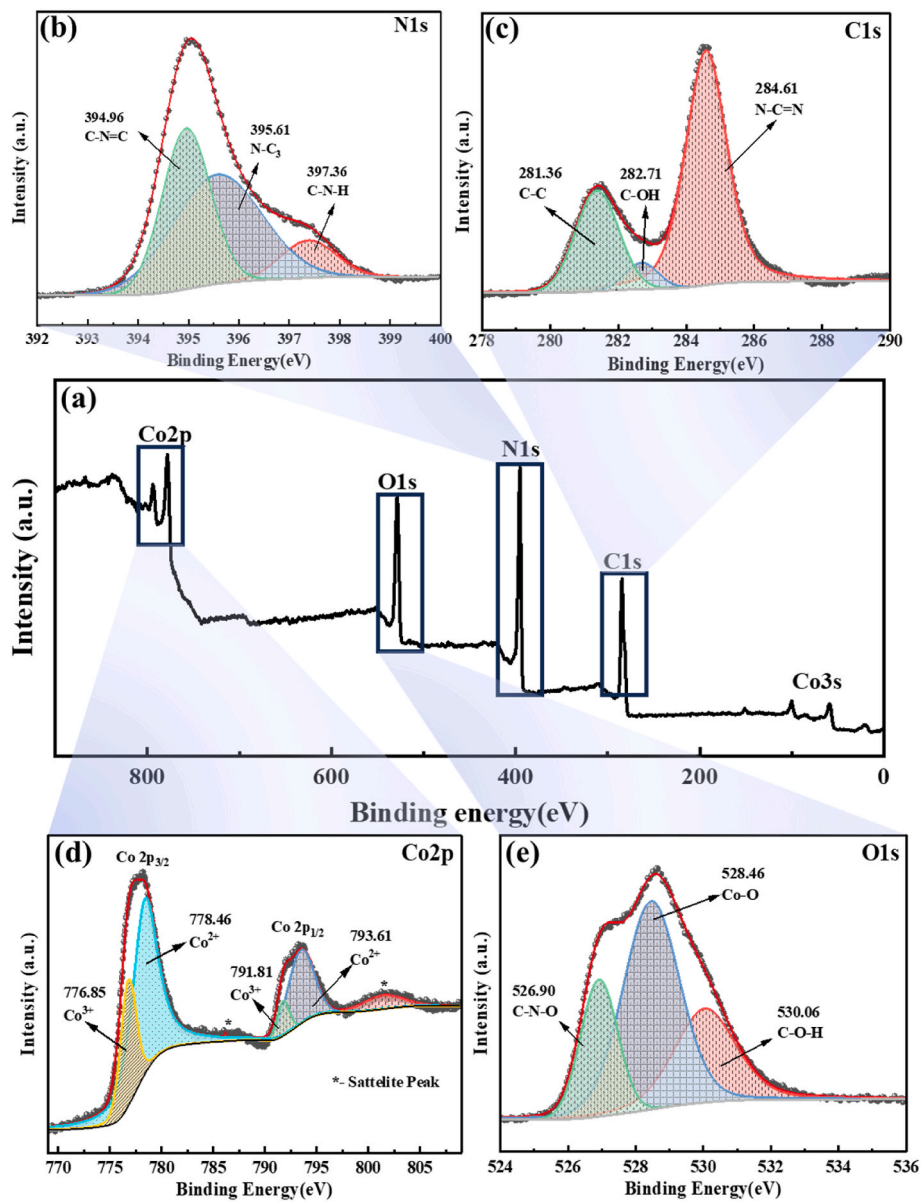


Fig. 6. (a) Survey spectra of COCN2, deconvoluted XPS spectra of (a) C1s, (b) N1s, (c) Co2p, and (e) O1s.

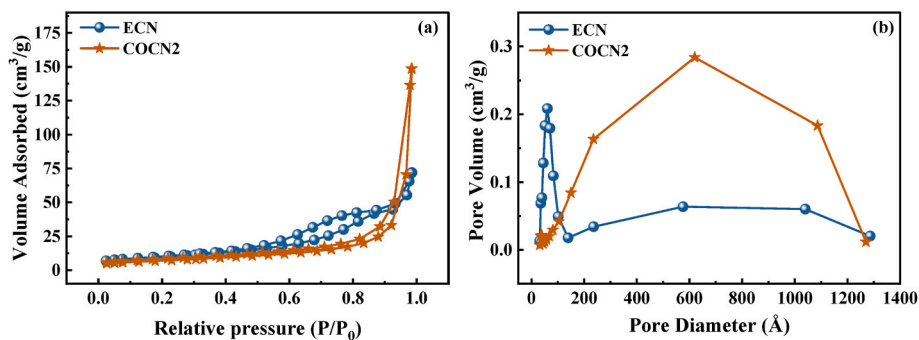


Fig. 7. (a) Adsorption and desorption isotherms of ECN and COCN2, (b) Pore size distribution of ECN, and COCN2.

adsorption extending beyond a single layer [45]. The ECN sample has an average pore size of about 60 Å and a specific surface area (SSA) of 25 m²/g. In contrast, the COCN2 sample features an average pore size of approximately 620 Å and an SSA of 35 m²/g. Both ECN and COCN2

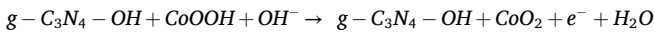
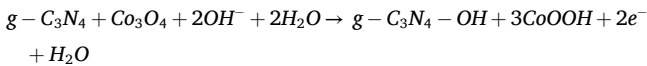
samples contain macropores ranging from 40 to 1200 Å in size. The COCN2 sample demonstrates higher capacitance due to the presence of macropores on the electrode surface, which facilitate the movement of electrolyte ions. The BET test results indicate that ECN and COCN2 are

well-suited for diffusion processes in supercapacitor applications, owing to their excellent SSA and average pore size [46].

3.7. Electrochemical analysis

3.7.1. Three-electrode electrochemical performance

Fig. 8(a) depicts the CV curves for ECN, Co_3O_4 , COCN1, COCN2, and COCN3 at a scan rate of 50 mV/s, spanning a potential range from 0.0 to 0.8 V. Notably, the COCN2 nanocomposite exhibits a larger area enclosed by its CV curve compared to ECN/ Co_3O_4 and the other COCN nanocomposites, indicating improved capacitive performance. The synergy between ECN and Co_3O_4 leads to an expanded CV integral area, highlighting COCN2's superior capacitive capabilities. Fig. 8(b) shows the CV plot for the COCN2 sample at various scan rates ranging from 2 to 100 mV/s within the same potential window. Interestingly, as the scan rate increased, the CV curves largely retained their shape, with only minor shifts in the positions of the redox peaks. This observation suggests that increased electrode polarization is associated with enhanced response kinetics in the electrode material, potentially resulting in higher current responses and an increase in surface redox reactions [47]. The CV curves for ECN, Co_3O_4 , COCN1, COCN2, and COCN3 demonstrate similar trends. Each sample features a CV curve that signifies an electrochemical reaction predominantly governed by redox mechanisms. The peaks that arise during the scanning process are due to faradaic reactions at the electrode. Interestingly, the two oxidation peaks at 0.58 V and 0.63 V represent the oxidation processes of cobalt's distinct oxidation states (Co^{2+} and Co^{3+}) [24]. The following section provides a detailed explanation of the observed redox reactions.:



The C_{sp} of each sample was determined using the equation below [48], and Fig. 8(c) compares the C_{sp} of each sample at the same SR of 2 mV/s.

$$C_s = \frac{\int I.dV}{2 \times m \times \vartheta \times \Delta V} \quad (1)$$

The loaded mass of the electrode is represented by m , ϑ stands for SR, ΔV for the applied PW, and $\int I.dV$ is the area under the CV plot. At a scan rate of 2 mV/s, the C_{sp} values for ECN, Co_3O_4 , COCN1, COCN2, and COCN3 are determined to be 62.52 F/g, 100.52 F/g, 168.27 F/g, 242.57 F/g, and 222.71 F/g, respectively. However, as the SR increases, there is a noticeable decline in C_{sp} values. This reduction is due to the limited time available for hydroxide ions (OH^-) in the electrolyte solution to penetrate the inner active sites of the electrode materials, which becomes more constrained as the SR rises. Therefore, the decreased scanning speeds allowed for the complete engagement of all active reaction centres within the electrode material [49]. This suggests that at lower SRs, a greater number of surface active sites are accessible for effective redox reactions compared to higher SRs. In CV measurements, two predominant electrochemical processes usually occur at the electrode: the capacitive electric double-layer process and the diffusion-controlled faradaic process. The interplay between these processes is characterized by a power law equation [50].

$$i(V) = a\vartheta^b \quad (2)$$

The variables a and b hold significant importance, with b specifically representing the charge storage mechanism within the electrode. A b value of 0.5 signifies diffusion-controlled redox reactions, whereas a b value of 1 indicates surface-controlled capacitance processes. When b is between 0.5 and 1, the electrochemical reactions involve a mix of diffusion-controlled redox reactions and surface-controlled capacitive processes. Fig. 8(d) displays a graph plotting $\log(i)$ versus $\log(\vartheta)$ for the materials ECN, Co_3O_4 , COCN1, COCN2, and COCN3. The analysis of these graphs yielded slope values that correspond to b values of 0.75, 0.56, 0.65, 0.73, and 0.68 for ECN, Co_3O_4 , COCN1, COCN2, and COCN3, respectively. These findings suggest that the nanocomposites utilize a blend of diffusion-controlled and surface-controlled mechanisms for storing charge. To further understand the mechanisms of charge storage at the electrode during electrochemical experiments, the current at the peak potential was extracted from CV graphs at various SRs. This

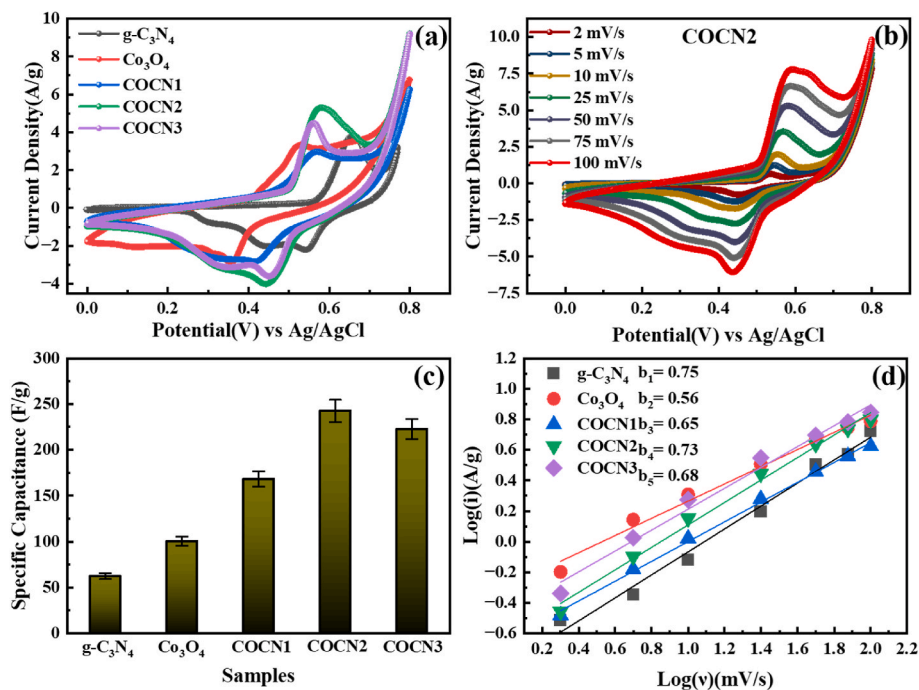


Fig. 8. (a) CV comparison of ECN, Co_3O_4 , COCN1, COCN2 and COCN3 at 50 mV/s SR, (b) CV plot of COCN2 at different SR from 2 to 100 mV/s, (c) Specific capacitance comparison of ECN, Co_3O_4 , COCN1, COCN2, and COCN3 at 2 mV/s SR, (d) A plot of $\log(i)$ vs $\log(v)$ of ECN, Co_3O_4 , COCN1, COCN2, and COCN3.

method enabled the identification and quantification of the charge storage contributions within the electrodes. The total current is composed of contributions from two distinct electrochemical processes. Dunn proposed the following equations, Eq. (3) and Eq. (4), as the corresponding representations [10].

$$i(V)_{total} = i(V)_c + i(V)_d \quad (3)$$

$$i(V) = k_1 v + k_2 v^{1/2} \quad (4)$$

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

$i(V)_c$ and $i(V)_d$ are currents for surface-controlled and diffusion-controlled processes, respectively. Using the equations above, graphing the $i(V)/v^{1/2}$ against the $v^{1/2}$ yields the slope (k_1) and intercept of the Y axis (k_2). Fig. 9(a, b, c, d, and e) illustrates the percentage of faradaic processes that are either governed by surface interactions or diffusion, which together contribute to the electrode's total capacity. As the SR increases, there is a clear decrease in the role of diffusion-controlled processes, while surface-controlled redox processes become more prominent. This indicates that electrolyte intercalation takes place at the interface where the electrode and electrolyte meet. The overall current results from a combination of surface-driven and diffusion-driven processes, both of which are crucial for determining the electrode's performance.

The GCD plots served as a tool to assess the electrochemical properties of the ECN, Co_3O_4 , COCN1, COCN2, and COCN3 electrodes, focusing on their charge storage potential and performance at varying rates. Fig. 10(a) presents a comparison of the discharging times for the ECN, Co_3O_4 , COCN1, COCN2, and COCN3 electrodes at a CD of 2.0 A/g. The graph clearly indicates that the COCN2 nanocomposite electrode exhibits the longest discharging time. Since discharging time is directly linked to C_{sp} , it is expected that the COCN2 nanocomposite electrode will have the highest C_{sp} . Consequently, the GCD of the COCN2 nanocomposite electrode was measured within a potential window of 0.0–0.7 at current densities ranging from 0.5 to 10 A/g, as depicted in Fig. 10(b). The shape of the charge-discharge curves further confirmed the pseudocapacitive behavior of all electrodes. As previously mentioned, the superior electrochemical performance of the nanocomposites is attributed to their increased number of active sites and porous structure, which enhances electrolyte diffusion. The C_{sp} for all samples was calculated using the discharging time and the formula

provided below [51].

$$C_{sp} = \frac{I \times \Delta t}{m \times \Delta V} \quad (5)$$

In this context, I denotes the applied current, Δt signifies the time taken for discharge, m represents the mass loaded, and ΔV stands for the potential difference. Fig. 10(c) presents a comparison of the C_{sp} for each sample at a uniform CD of 0.5 A/g. The C_{sp} values for ECN, Co_3O_4 , COCN1, COCN2, and COCN3 are recorded as 80.02 F/g, 173.153 F/g, 404.641 F/g, 667.85 F/g, and 537.5 F/g, respectively, at a CD of 0.5 A/g, respectively. Fig. 10(d) illustrates a Ragone plot, a graphical representation used to compare the ED and PD of various energy storage systems. This figure highlights the Ragone plots for each sample, revealing the variations in their ED and PD. The ED and PD values for these samples were determined using the equations below, which are based on the GCD plot [52].

$$\text{Energy Density (ED)} = \frac{C_s \Delta V^2}{2} \quad (6)$$

$$\text{Power Density (PD)} = \frac{ED}{\Delta t} \quad (7)$$

here, ΔV represents the potential difference applied, while Δt denotes the time taken for discharging. When comparing ED to PD, the COCN2 nanocomposite achieves the highest values, reaching 45.45 Wh/Kg and 218 W/kg. The notable PD and ED of the COCN2-based electrode material indicate rapid charging and discharging capabilities, along with adequate energy storage. Stability of the electrode material is essential for potential energy storage applications. Despite the COCN2 electrode demonstrating the best capacitive efficiency among all tested electrodes, it was the only one subjected to an electrode stability test. Fig. 10(e) illustrates the results from analyzing the COCN2 electrode through repeated charge-discharge cycles. Electrochemical stability assessments were performed on COCN2 electrodes at varying current densities of 2.0 A/g, 10 A/g, 15 A/g, and 20 A/g to evaluate the performance of the electrode under higher current conditions. Assessing its cyclic performance at current densities of 2.0 A/g, 10 A/g, 15 A/g, and 20 A/g, the COCN2 electrode exhibited a capacitive retention of around 99.15 %, 99.25 %, 98.15 %, and 96.87 % following 1000 continuous cycles. These results suggest that the NCOCN2 electrode has good electrochemical retentivity at higher current densities. The C_{sp} and ED values are compared with those reported in previous studies in Table 1 [53–56].

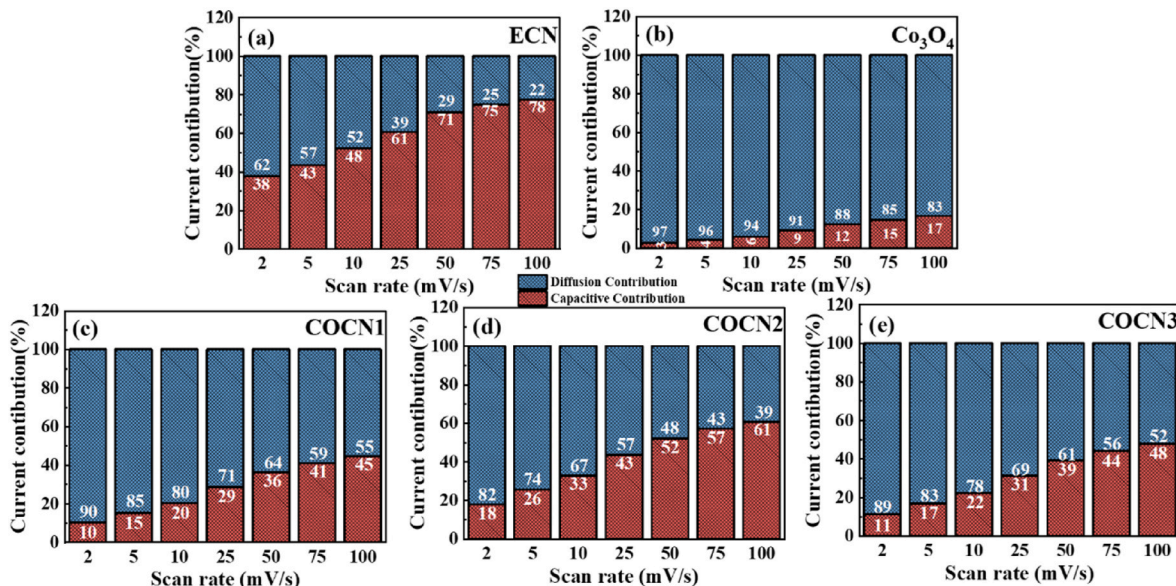


Fig. 9. Capacitive and diffusion contribution plots of (a) ECN, (b) Co_3O_4 , (c) COCN1, (d) COCN2, and (e) COCN3.

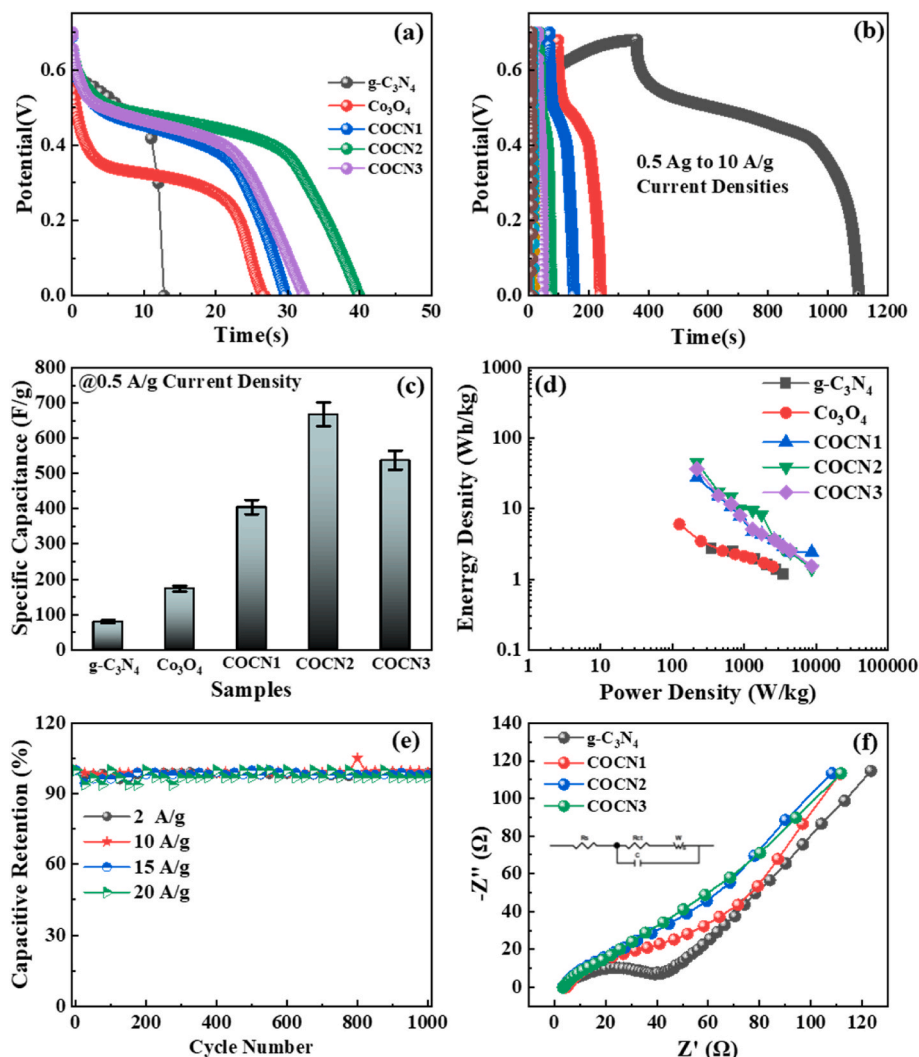


Fig. 10. (a) Discharging time comparison of ECN, Co_3O_4 , COCN1, COCN2, and COCN3, (b) GCD plot of COCN2 at 0.5–10 A/g CDs, (c) Specific capacitance comparison of ECN, Co_3O_4 , COCN1, COCN2, and COCN3, (d) Ragone plot of ECN, Co_3O_4 , COCN1, COCN2, and COCN3, (e) Stability test of COCN2 electrode at different current densities, (f) Nyquist plot of ECN, COCN1, COCN2, COCN3 with equivalent circuit.

Table 1

The comparison of specific capacitance, energy density, power density, and cycling stability of COCN2 nanocomposites with previous study.

Electrode	Specific Capacitance @ Current Density	Energy Density @ Power Density	Electrolyte	Capacitive Retention @No. of Cycles)	Ref.
Co_3O_4 /graphite	395.04 F/g @ 0.5 A/g	–	6 M KOH	97.3 % @ 1000 cycles	[53]
graphene oxide/ Co_3O_4	458 F/g @ 0.5 A/g	39.0 Wh/kg @ 8300 W/kg	2 M KOH	95.6 % @ 1000 cycles	[54]
$\text{ZnO/g-C}_3\text{N}_4$	85.47 F/g @ 1 A/g	24 Wh/kg @ 750 W/kg	6 M KOH	82.1 % @ 3000 cycles	[55]
$\text{CuO@g-C}_3\text{N}_4$	98 F/g @ 1 A/g	14.8 Wh/kg @ 988.16 W/kg	1 M KOH	81 % @ 2500 cycles	[56]
$\text{g-C}_3\text{N}_4/\text{Co}_3\text{O}_4$ (COCN2)	667.8 F/g @ 0.5 A/g	45.45 Wh/kg @ 218.75 W/kg	1 M KOH	99.5 % @ 1000 cycles	This Work

To gain a deeper insight into the electrode materials used in supercapacitors, the impedance profiles of ECN, COCN1, COCN2, and COCN3 were examined. As depicted in Fig. 10(f), the electrodes were assessed at an open circuit voltage (OCV) across a frequency range from 0.1 Hz to 100 kHz. The Nyquist plots displayed a slanted line in the low-frequency area and a semicircular section in the high-frequency area. In the high-frequency domain, the solution resistance (R_s) forms at the interface between the electrode and electrolyte, whereas in the low-frequency domain, the charge transfer resistance develops on the electrode's surface [57]. The resistance to electron flow at the electrode/electrolyte interface is directly related to the diameter of the semicircle. As illustrated in Fig. 10(f), the R_s and charge transfer resistance (R_{ct}), Warburg impedance coefficient (W_s), and double layer capacitance (C) values for

all electrodes were utilized to analyze their experimental and fitted AC impedance graphs. The equivalent circuit model comprises R_s , R_{ct} , W_s , and C . The Zview program was employed to determine the equivalent circuit and its values, shown in Table 2 [50]. The constant phase element is used to calculate the double-layer capacitance and low-frequency

Table 2

The comparison of R_s , R_{ct} , W_s , and C of gCN, COCN1, COCN2, and COCN3.

Sample Name	$R_s(\Omega)$	$R_{ct}(\Omega)$	$W_s(\Omega/s^{0.5})$	C (mF)
ECN	4.48	28.1	107.2	80.32
COCN1	5.90	26.84	87.67	108.9
COCN2	4.16	12.41	80.06	157.2
COCN3	4.37	19.11	70.98	75.37

pseudo-capacitance. The R_s values for ECN, COCN1, COCN2, and COCN3 were measured at 4.82 Ω , 5.91 Ω , 4.16 Ω , and 4.37 Ω , respectively. A lower R_s value corresponds to the behavior of an ideal capacitor. The R_{ct} values for ECN, COCN1, COCN2, and COCN3 were recorded as 40.3 Ω , 26.84 Ω , 12.41 Ω , and 19.11 Ω , respectively. Low R_s and R_{ct} values allow the electrode material to exhibit high conductivity, thereby improving its overall performance. The W_s value for ECN, COCN1, COCN2, and COCN3 were determined as 107.2, 87.67, 80.06, and 70.98, respectively. Among all samples, COCN3 exhibits the lowest Warburg impedance (70.98 $\Omega/s^{0.5}$), indicating improved ion diffusion, while COCN2 shows the highest double-layer capacitance (157.2 mF), reflecting superior charge storage capability at the electrode–electrolyte interface. Consequently, the NCOCN2 nanocomposite shows overall good electrochemical performance, so it is suitable for use in supercapacitors.

3.7.2. Electrochemical analysis of COCN2-based symmetric supercapacitor device with Whatman paper as a separator (COCN2/W/COCN2 SS CD)

The electrochemical evaluation of the ECN, COCN1, COCN2, and COCN3 nanocomposites, conducted using a three-electrode configuration, revealed that COCN2 exhibited the most favourable electrochemical properties. This was attributed to the synergistic interaction between the layered ECN and Co_3O_4 nanoparticles, which significantly enhanced the supercapacitor's overall performance, particularly in

terms of rate capability and increased capacitance. As a result, COCN2 was chosen for the development of a SS CD. This device was constructed by applying equal amounts of COCN2 on both sides, with a Whatman filter paper soaked in a 1.0 M KOH electrolyte solution placed in the center, all arranged using a Swagelok setup. The CV plot of the COCN2/W/COCN2 SS CD at a scan rate of 200 mV/s is depicted across several potential windows in Fig. 11(a). Even at a scan rate of 200 mV/s, the SS CD maintains its impressive electrochemical performance. It is important to highlight that the SS CD exhibits pseudocapacitive behavior across all potential windows and consistently shows a redox peak. The experimental peak is attributed to the presence of Co_3O_4 and ECN within the COCN2 nanomaterial. The graph illustrates multiple oxidation and reduction processes that occur due to redox reactions involving ions from the KOH electrolyte. The high nitrogen content of ECN and the multiple oxidation states of Co_3O_4 facilitate these redox reactions [27]. The COCN2/W/COCN2 SS CD operates optimally within a potential window of -1.6 to $+1.6$ V. A CV plot of COCN2/W/COCN2 SS CD assessment at various scan rates ranging from 5 mV/s to 100 mV/s is shown in Fig. 11(b) within the potential window of -1.6 V to $+1.6$ V. As previously mentioned, the COCN2 nanocomposites possess a porous structure with multiple active sites that enhance ion transfer and enable electrolyte absorption. Therefore, the COCN2/W/COCN2 SS CD can function at different scan rates and potential windows. The C_{sp} of COCN2/W/COCN2 SS CD was calculated using a CV graph, and the C_{sp} versus scan

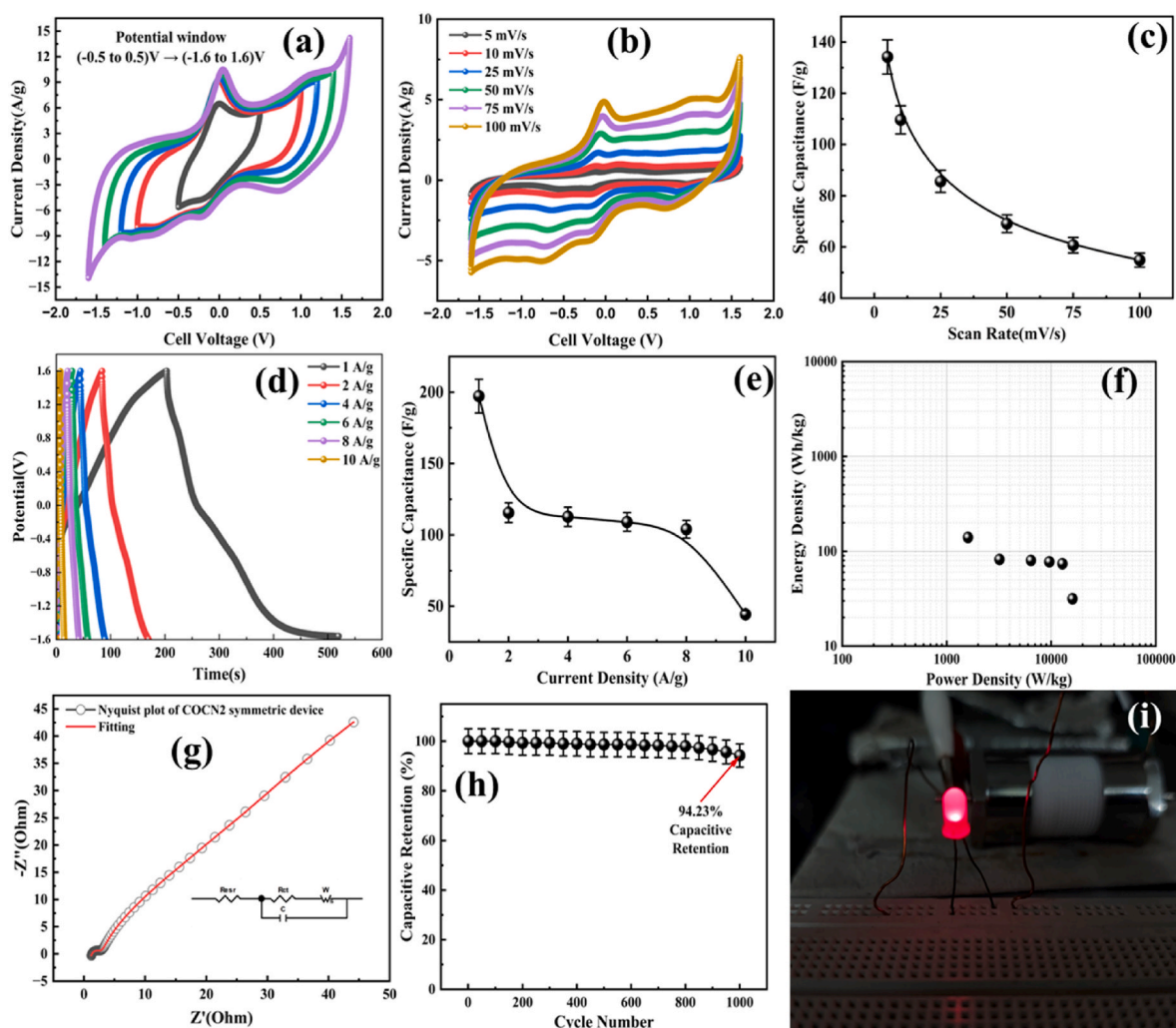


Fig. 11. (a) CV of COCN2/W/COCN2 SS CD at different PWs, (b) CV of COCN2/W/COCN2 SS CD at different SR, (c) Specific capacitance vs SR of COCN2/W/COCN2 SS CD, (d) GCD plot of COCN2/W/COCN2 SS CD at different CDs, (e) Specific capacitance vs CDs plot of COCN2/W/COCN2 SS CD, (f) Ragone plot of COCN2/W/COCN2 SS CD, (g) Nyquist plot of COCN2/W/COCN2 SS CD with equivalent circuit, (h) Stability test of COCN2/W/COCN2 SS CD, (i) Picture of LED glow by SS CD.

rate curve is shown in Fig. 11(c). The COCN2/W/COCN2 SSCD achieves a maximum C_{sp} of 134 F/g at a scan rate of 5 mV/s. The C_{sp} value decreases as the scan rate increases because, at higher scan rates, ions have less time to penetrate the electrode material, which affects the charge storage process [58].

Fig. 11(d) illustrates the evaluation of GCD curves at different CDs ranging from 1.0 A/g to 10.0 A/g within the optimal PW. The shape of these curves once again verifies the pseudocapacitance characteristics of the device. The C_{sp} of a COCN2/W/COCN2 SSCD was calculated using a GCD graph, and Fig. 11(e) presents a graph of C_{sp} values against CDs. At a CD of 1 A/g, the COCN2/W/COCN2 SSCD achieves a peak C_{sp} of 197.25 F/g. Notably, even at higher CDs like 10 A/g, capacitive behavior is observed, indicating that the COCN2/W/COCN2 SSCD meets the requirements for energy storage applications. Fig. 11(f) employs the Ragone plot method to depict the PD and ED of the COCN2/W/COCN2 SSCD, which delivers a maximum PD of 1600 W/kg and an ED of 140.27 Wh/kg. This data highlights the rapid charge and discharge capabilities of the COCN2/W/COCN2 SSCD.

Fig. 11(g) presents the Nyquist impedance plot along with the corresponding circuit fit for the device. The measurements span a frequency range from 100 kHz to 0.1 Hz, with an OCV of +0.005 V. In the Nyquist plot, a linear trend is observed at low frequencies, which does not perfectly align with the Y-axis. This characteristic is attributed to the rough and porous nature of the COCN2 nanocomposite, facilitating optimal electrolyte penetration and enhancing ion diffusion efficiency [59]. The x-intercept in the high-frequency region represents the

equivalent series resistance (R_{esr}), which is the total resistance from both the electrode and electrolyte. By fitting the equivalent circuit of the COCN2/W/COCN2 SSCD, the R_{esr} and R_{ct} values are found to be 0.83 Ω and 1.84 Ω , respectively. These low R_{esr} and R_{ct} values align with the GCD results, indicating minimal internal resistance and a swift charging-discharging process [31]. To evaluate the durability of the COCN2/W/COCN2 SSCD, the device undergoes 1000 continuous charging-discharging cycles at a CD of 5.0 A/g, with the capacitive retention graph depicted in Fig. 11(h). The COCN2/W/COCN2 SSCD demonstrates excellent charging-discharging performance, maintaining 94.23 % of its initial capacitance. For practical application, the COCN2/W/COCN2 was connected to a RED LED, which it successfully illuminated, as shown in Fig. 11(i).

3.7.3. Electrochemical analysis of COCN2 based symmetric supercapacitor device with gel electrolyte as a separator (COCN2/GE/COCN2 SSCD)

To investigate the impact of a separator on the electrochemical performance of the COCN2-based SSCD, a PVA-KOH gel electrolyte was employed as both separator and electrolyte. This approach offers benefits such as reduced leakage, enhanced stability and flexibility, improved interface resistance, and high ionic conductivity. The electrochemical performance of the COCN2/GE/COCN2 SSCD device was evaluated using CV, GCD, and EIS techniques. The CV plot for the COCN2/GE/COCN2 SSCD was recorded at a SR of 200 mV/s across different PWs to check the optimal operating PW to run the device, as shown in Fig. 12(a). Remarkably, even at this high SR, the SSCD

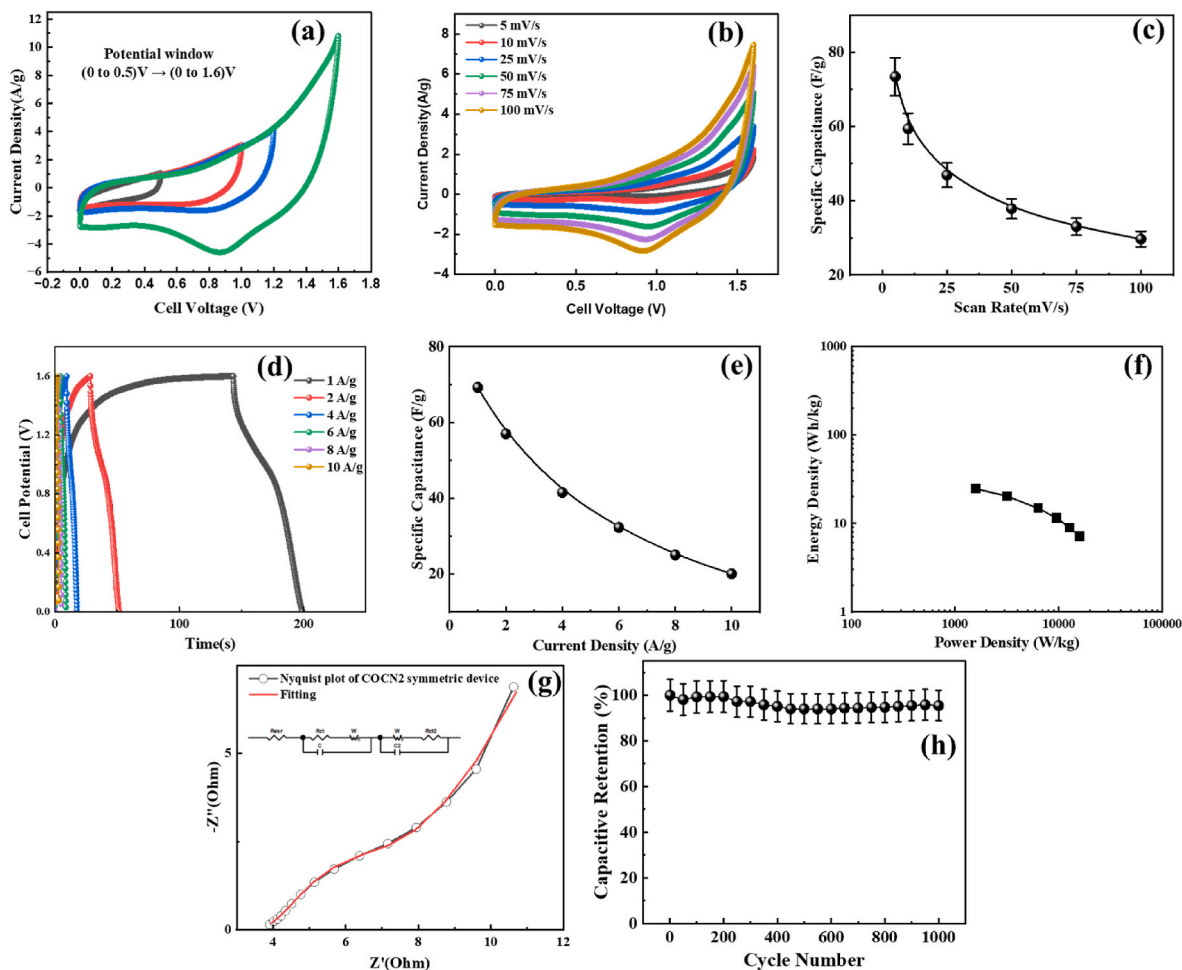


Fig. 12. (a) CV of COCN2/GE/COCN2 SSCD at different PWs, (b) CV of COCN2/GE/COCN2 SSCD at different SR, (c) Specific capacitance vs SR of COCN2/GE/COCN2 SSCD, (d) GCD plot of COCN2/GE/COCN2 SSCD at different CDs, (e) Specific capacitance vs CDs plot of COCN2/GE/COCN2 SSCD, (f) Ragone plot of COCN2/GE/COCN2 SSCD, (g) Nyquist plot of COCN2/GE/COCN2 SSCD with equivalent circuit, (h) Stability test of COCN2/GE/COCN2 SSCD.

demonstrates excellent electrochemical performance. It is noteworthy that the device exhibits pseudocapacitive behaviour throughout the entire PW, consistently displaying a redox peak. This peak is attributed to the presence of Co_3O_4 and ECN within the COCN2 nanomaterial. As previously discussed in the electrochemical analysis of the COCN2/W/COCN2 SSCD, these redox peaks result from oxidation and reduction processes driven by reactions involving ions from the PVA-KOH gel electrolyte. These reactions are facilitated by the high nitrogen content in ECN and the dual oxidation states of cobalt (Co^{2+} and Co^{3+}) in the Co_3O_4 component of the COCN2 nanocomposite. Based on these observations, the optimal operating PW for the device was determined to be between 0 V and +1.6 V. Fig. 12(b) presents the CV plot of the COCN2/GE/COCN2 SSCD evaluated at different SRs, ranging from 5 mV/s to 100 mV/s, within the PW of 0 V to +1.6 V. As previously noted, the highly porous structure of the COCN2 nanocomposite, enriched with numerous active sites, enhances ion and electron transport within the supercapacitor device and ensures efficient electrolyte absorption [31, 32]. This enables the COCN2/GE/COCN2 SSCD to operate effectively across a wide range of SRs and PWs. At higher SRs, a noticeable increase in current response was observed, consistent with previously reported data. The C_{sp} of the COCN2/GE/COCN2 SSCD was calculated using the CV data, and the relationship between C_{sp} and SR is illustrated in Fig. 12(c). The device achieves a maximum C_{sp} of 73.4 F/g at a SR of 5 mV/s. However, as the SR increased, the C_{sp} decreased. This trend occurs because higher SRs alter the charge storage mechanism, limiting the ability of electrolyte ions to access the active sites on the electrode surface. In contrast, at lower SRs, electrolyte ions have more time to reach these active sites, resulting in higher C_{sp} values.

As depicted in Fig. 12(d), these curves were obtained at varying currents. The GCD curve profiles reveal rapid charging and discharging cycles, highlighting the device's outstanding electrochemical capacitance properties and confirming its pseudocapacitive nature. From the GCD data, the C_{sp} of the COCN2/GE/COCN2 SSCD was determined. Fig. 12(e) illustrates the correlation between C_{sp} and CD. At a CD of 1 A/g, the device reaches a peak C_{sp} of 69.25 F/g. Importantly, the device demonstrated its suitability for energy storage applications by retaining its capacitive behaviour even at high CDs, such as 10 A/g. Additionally, the ED and PD of the COCN2/GE/COCN2 SSCD were assessed using a Ragone plot, as shown in Fig. 12(f). The COCN2/GE/COCN2 SSCD achieves a maximum ED and PD of 24.62 Wh/kg and 1600 W/kg, respectively, indicating its capability to efficiently store and deliver energy. By comparing this result with COCN2/W/COCN2, it is evident that the device utilising Whatman paper exhibits higher C_{sp} , ED, and PD. This is attributed to the more porous nature of Whatman paper, which allows the liquid KOH electrolyte to soak in more effectively, resulting in a more uniform and well-wetted electrode surface. A greater amount of electrolyte facilitates increased ion accessibility to the electrode, thereby enhancing electrochemical performance. Table 3 provides a comparison of both SSCD with previously reported studies [60–62]. The electrical impedance of the COCN2/GE/COCN2 SSCD was evaluated using EIS, with the results presented in a Nyquist plot, as depicted in Fig. 12(g). The R_{esr} for the COCN2/GE/COCN2 SSCD was measured at 3.693 Ω , suggesting minimal internal resistance to ion transport. The R_{esr} value of the COCN2/GE/COCN2 is higher than that of the COCN2/W/COCN2 SSCD. This suggests that using Whatman paper reduces the ESR resistance, thereby improving the electrochemical performance of the SSCD. The COCN2/GE/COCN2 SSCD exhibits two charge transfer resistances, R_{ct1} and R_{ct2} , which are 1.636 Ω and 0.07 Ω , respectively. This indicates efficient charge kinetics between the electrolyte and electrode material in the COCN2/GE/COCN2 SSCD. The nearly vertical trend in the post-semicircle region could be attributed to the porous structure of the COCN2 nanocomposite and the effective ion transport facilitated by a gel electrolyte separator [32,63]. The inclusion of a gel electrolyte enhances the stability and ion accessibility of the COCN2/GE/COCN2 SSCD. The COCN2/GE/COCN2 SSCD underwent 1000 continuous charging-discharging cycles at a CD of 5.0 A/g to

Table 3

The comparison of specific capacitance, energy density, power density, and cycling stability of symmetric supercapacitor device with previous study.

Symmetric supercapacitor device	Specific Capacitance/ Capacity @ Current Density	Energy Density @ Power Density	Separator	Capacitive Retention @No. of Cycles	Ref.
$\text{Co}_3\text{O}_4/\text{rGO}/\text{NF}$	311 F/g @ 1 A/g	62.2 Wh/kg @ 599.84 W/kg	Filter paper	89.69 % @ 10000 cycles	[61]
$\text{Co}_3\text{O}_4/\text{g-C}_3\text{N}_4$	125 F/g @ 0.5 A/g	28.13 Wh/kg @ 337.5 W/kg	PEO/PEGDME/KOH/ based gel electrolyte	90 % @ 5000 cycles	[25]
$\text{Bi}_2\text{S}_3/\text{g-C}_3\text{N}_4$	25 $\mu\text{Ah}/\text{cm}^2$ @ 1 mA/cm^2	3.17 $\mu\text{W h}/\text{cm}^2$ @ 1495 $\mu\text{W}/\text{cm}^2$	PVA-KOH gel electrolyte	83.84 % @ 7500 cycles	[62]
COCN2/W/COCN2	197.25 F/g @ 1 A/g	140 Wh/kg @ 1600 W/kg	Whatman paper	94.23 % @ 1000 cycles	This Work
COCN2/GE/COCN2	69.25 F/g @ 1 A/g	24.62 Wh/kg @ 1600 W/kg	PVA-KOH gel electrolyte	95.4 % @ 1000 cycles	This Work

evaluate its long-term durability. The capacitive retention graph is shown in Fig. 12(h). The COCN2/GE/COCN2 SSCD exhibits excellent charging-discharging performance and retains 95.4 % of its initial capacitance. The COCN2/GE/COCN2 used SSCD has better retention than the COCN2/W/COCN2. This is attributed to a more stable structure provided by the polymer matrix of PVA-KOH GE, which minimises electrolyte leakage, evaporation and degradation over time, leading to better long-term stability. Following the assessment of long-term stability, the morphology was examined using SEM for the COCN2 electrode in its prepared state and after cycling, as illustrated in Fig. 13(a and b), respectively. This image distinctly illustrates the minimal morphological degradation, indicating the electrode's strong performance after 1000 GCD cycles. Fig. 13(c and d) illustrates the EDS spectrum of the as-prepared electrode and the electrode after cycling, showcasing the elements present in this electrode. According to all of these findings, the $\text{Co}_3\text{O}_4/\text{g-C}_3\text{N}_4$ nanocomposite may find usage as a supercapacitor in energy storage applications.

4. Conclusion

The ECN/ Co_3O_4 nanocomposites were synthesized using the ultrasonication wave method. XRD analysis identified a cubic spinel structure for Co_3O_4 and a hexagonal crystal structure for ECN. The crystallite sizes for Co_3O_4 , COCN1, COCN2, and COCN3 were determined to be 10 nm, 9 nm, 6.9 nm, and 6.8 nm, respectively. Correspondingly, the lattice strain values were calculated as 1.03×10^{-2} , 1.21×10^{-2} , 2.17×10^{-2} , and 2.23×10^{-2} . The decrease in crystallite size along with the increase in lattice strain indicates that the COCN nanocomposites, especially COCN2 and COCN3, possess optimized structural features. FESEM analysis showed that the COCN nanocomposites have a porous and rough texture. XPS analysis suggests that the presence of multiple oxidation states of cobalt (Co^{2+} and Co^{3+}) in Co_3O_4 and the high nitrogen content in ECN enhance the redox activities of the nanocomposites in electrochemical properties. Among all the samples, COCN2 nanocomposites demonstrated a high C_{sp} of 667.8 F/g at 0.5A/g and retained 99.5 % of their capacitive performance after 1000 charge-

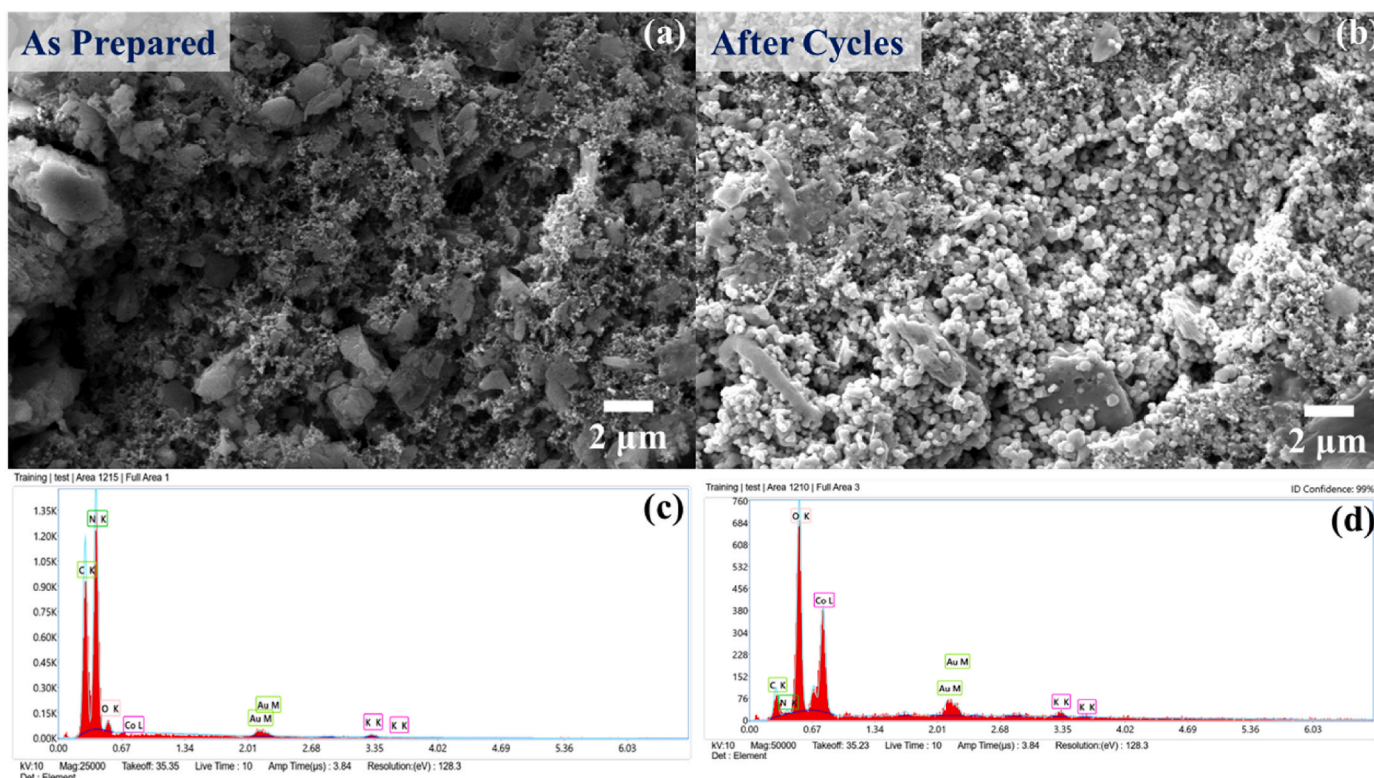


Fig. 13. SEM images of NCOCN2 electrode (a) As prepared, (b) After cycle; EDS spectra of (a) As prepared, (b) After cycle.

discharge cycles. The COCN2/W/COCN2 and COCN2/GE/COCN2 SSCDs achieved a C_{sp} of 197.25 F/g and 69.25 F/g at 1.0 A/g, respectively. Among these SSCDs, COCN2/W/COCN2 exhibited a high ED of 140.27 Wh/kg at 1600 W/kg with 94.23 % capacitive retention after 1000 charge-discharge cycles. Consequently, it shows that Whatman paper-based SSCD outperforms PVA-KOH gel electrolyte-based SSCD in terms of C_{sp} , ED, and PD. These findings indicate that COCN nanocomposites hold significant promise for conventional energy storage applications due to their optimized structural properties, porous morphology, multiple oxidation states, and favourable surface area.

CRediT authorship contribution statement

Manas Nasit: Writing – original draft, Investigation, Formal analysis, Data curation. **Kavita Kumari:** Writing – review & editing, Resources, Formal analysis. **Naveen Yadav:** Resources, Formal analysis. **Bon-Heun Koo:** Writing – review & editing, Validation, Resources, Formal analysis. **Saurabh Dalela:** Writing – review & editing, Validation. **Ankush Vij:** Writing – review & editing, Supervision, Formal analysis. **Aditya Sharma:** Visualization, Formal analysis. **P.A. Alvi:** Validation, Formal analysis. **Shalendra Kumar:** Writing – review & editing, Supervision, Formal analysis, Conceptualization.

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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