



Article

Flower-Shaped CoS-Co₂O₃/G-C₃N₄ Nanocomposite for Two-Symmetric-Electrodes Supercapacitor of High Capacitance Efficiency Examined in Basic and Acidic Mediums

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Abstract: Graphitic carbon nitride (G-C₃N₄) was synthesized through the direct combustion of urea in the air. The CoS-Co₂O₃/G-C₃N₄ composite was synthesized via the hydrothermal method of G-C₃N₄ using cobalt salts. The morphological and chemical structures were determined through XRD, XPS, SEM, and TEM. XRD and XPS analyses confirmed the chemical structure, function groups, and elements percentage of the prepared nanocomposite. SEM measurements illustrated the formation of G-C₃N₄ sheets, as well as the flower shape of the CoS-Co₂O₃/G-C₃N₄ composite, evidenced through the formation of nano appendages over G-C₃N₄ sheets. TEM confirmed the 2D nanosheets of G-C₃N₄ with an average width and length of 80 nm and 170 nm, respectively. Two symmetric electrodes for the supercapacitor from the CoS-Co₂O₃/G-C₃N₄ composite. Electrochemical measurements were carried out to determine the charge/discharge, cyclic voltammetry, stability, and impedance of the prepared supercapacitor. The measurements were carried out under acid (0.5 M HCL) and basic (6.0 M NaOH) mediums. The charge and discharge lifetime values in the acid and base medium were 85 and 456 s, respectively. The cyclic voltammetry behavior was rectangular in a base medium for the pseudocapacitance feature. The supercapacitor had 100% stability retention up to 600 cycles; then, the stability decreased to 98.5% after 1000 cycles. The supercapacitor displayed a specific capacitance (C_S) of 361 and 92 F/g, and an energy density equal to 28.7 and 30.2 W h kg^{−1} in the basic and acidic mediums, respectively. Our findings demonstrate the capabilities of supercapacitors to become an alternative solution to batteries, owing to their easy and low-cost manufacturing technique.

Keywords: supercapacitor; cobalt sulfide; cobalt oxide; graphitic carbon nitride; composite



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1. Introduction

Sustainable energy has become a promising field owing to cost-effective energy sources. Supercapacitors have been considered replacements for batteries. This is due to their simple charging, lasting a long time before being completely discharged [1,2]. Three types of supercapacitors, i.e., double-layer, hybrid, and pseudo-capacitors, have been developed so far. However, commercial supercapacitors are based on carbon materials (double-layer supercapacitors). Nevertheless, such capacitors have a large drawback related to the limited energy produced [3]. This has motivated researchers to develop advanced supercapacitors based on two composite carbon materials (hybrid supercapacitors). Indeed, the unique combination of composite materials ensures a better efficiency of supercapacitors that remains related to the redox reaction of the used material [4]. Particularly, the combination of carbon materials with redox materials in developing a supercapacitor ensures a significant enhancement of the capacitance through the charge storage inside the layers.

On the other hand, the electrochemistry of supercapacitors highly depends on the morphology of the used materials. For instance, using carbon layer materials decorated with additional metal oxides or sulfides increases the surface area that consequently originates extra active sites [5–8]. Consequently, this raises the charge storage inside the composite materials [3].

Graphitic carbon nitride (G-C₃N₄) is considered a promising material due to its simple preparation in a high-mass product, through the combustion of organic nitrogen materials such as urea and thiourea. G-C₃N₄ has great properties related to charge storage, represented by high surface area and charge chemical durability. This is assigned to the chemical structure of G-C₃N₄ of tris-triazine (C₆N₇) in the ternary amino group [9]. Santos et al. fabricated CoO and CuO decorated on G-C₃N₄; a specific capacitance (CS) of 124.75 and 84.28 at 0.5 A g were found, respectively [10]. Kuila et al. synthesized Gd decorated on G-C₃N₄ for supercapacitor applications; a capacitance equal to 2.59 mF cm^{−2} at 10 mV s^{−1} was obtained [11]. Furthermore, Rani et al. investigated the CoFe₂O₄/G-C₃N₄ composite, they demonstrated the impact of G-C₃N₄ on enhancing the electrical properties of the supercapacitor [12]. On the other hand, additional studies have been carried out on graphitic materials, such as mesoporous carbon nitride/graphene from 0.5 M H₂SO₄ as an electrolyte; a C_s of 240 F/g was produced [13]. Additionally, the C_s of G-C₃N₄/graphene electrode from 1 M KOH were studied and its value was equal to 265 F/g [14]. Similarly, a supercapacitor from 0.4 M LiClO₄ demonstrated a C_s of 264 F/g [15]. Additionally, G-C₃N₄/carbon nanotube was used as the supercapacitor from a polyvinyl alcohol/H₂SO₄ electrolyte; the produced C_s value was 148 F/g [16]. G-C₃N₄/activated carbon from 1 M Na₂SO₄; the produced C_s value was 266 F/g [17].

Herein, a flower-shaped CoS-Co₂O₃/G-C₃N₄ nanocomposite for a two-symmetric-electrodes supercapacitor is prepared and tested in base and acid mediums. All the chemical analyses for confirming the chemical structure and morphology are carried out. The electrochemical properties are tested by measuring the charge/discharge, cyclic voltammetry, impedance, and stability of the prepared supercapacitor. The supercapacitor demonstrates a promising electrical property in the basic medium in comparison to the acid medium. The supercapacitor has a great performance in comparison to instances in the literature.

2. Experimental Section

2.1. Materials

Urea, thiourea, HCl, NaCl, and NaOH were purchased from Piochem Co., Cairo, Egypt. While cobalt acetate (Co(CH₃COOH)₂) and ammonia (NH₄OH) were purchased from El Naser chemical Co., Cairo, Egypt. Nafion (5 wt.% dissolved in methanol) was obtained through Sigma–Aldrich, St. Louis, MO, USA.

2.2. Preparation of CoS-Co₂O₃/G-C₃N₄ Nanocomposite

First, 10 g of urea was well annealed in a tube furnace at 550 °C for 2 h under an N₂ atmosphere. This led to the combustion and conversion of urea to G-C₃N₄.

For the preparation of the composite, (0.1 M) Co(CH₃COOH)₂, (0.2 M) thiourea, and (0.5 M) urea were dissolved in 40 mL H₂O using an ultrasonic stirrer. Then, the pH solution was raised to 9 using an aqueous ammonia solution. Meanwhile, 0.05 g C₃N₄ was suspended in 30 mL H₂O. Then, both solutions were added to each other inside the autoclave, in which the hydrothermal reaction was carried out at 160 °C for 12 h. After that, the produced powder was collected, washed, and dried at 60 °C for 5 h. Finally, the powder was annealed at 300 °C for 10 min. This process caused the formation of the CoS-Co₂O₃/G-C₃N₄ nanocomposite.

2.3. Synthesis and Electrochemical Testing of the Supercapacitor

Synthesis of the symmetric supercapacitor was carried out using the main prepared materials CoS-Co₂O₃/G-C₃N₄ nanocomposite. This material was used as a paste by mixing

with (0.045 g) of nafion and 0.75 mL ethanol. Then, 0.003 g of this paste was loaded over an Au sheet (1 cm²). The supercapacitor was constructed by combining these two electrodes via a filter paper wetted with the electrolyte as a separator. Measurements were carried out under different electrolytes, such as HCl, NaOH, and NaCl, using an electrochemical workstation (CHI660E, USA) at 25 °C.

2.4. Characterization Process

The prepared materials were characterized using different analytical tools for confirming their chemical and morphological properties. X-ray diffraction (X'Pert Pro, Almelo Holland) and X-ray photoelectron spectroscopy were used to confirm the chemical structure of the prepared materials. The morphologies were determined using a scanning electron microscope, SEM, (ZEISS SUPRA 55 VP, Oberkochen, Germany) and a transmitted electron microscope (TEM), (JEOL JEM-2100).

3. Results and Discussion

The chemical and structural composition of the graphitic-like layer structures of G-C₃N₄ and CoS-Co₂O₃/G-C₃N₄ were confirmed using the XRD pattern (see Figure 1a). The G-C₃N₄ displayed two distinct diffraction peaks: a weak (100) diffraction peak at 13.03°, attributed to the in-planar structural packing motif with a separation of 0.679 nm, and a strong other peak located at 27.20° corresponding to the (002) peak of the interlayer d-spacing of 0.327 nm. The two peaks at 13.03° and 27.20° were matched with previous studies (JCPDS 87–1526 card) [18] and (JCPDS) 87–1526 for g-C₃N₄ [19]. The composite exhibited diffraction peaks corresponding to both g-C₃N₄ and CoS-Co₂O₃, reflecting the presence of two phases. The CoS nanomaterial had three peaks for the growth directions (101), (102), and (110), located at 32.45, 42.73, and 51.22°, respectively [20]. The Co₂O₃ nanomaterial had four peaks for the growth directions (311), (400), (422), and (440) for the positions 30.56, 38.51, 46.51, and 54.64°, respectively [21].

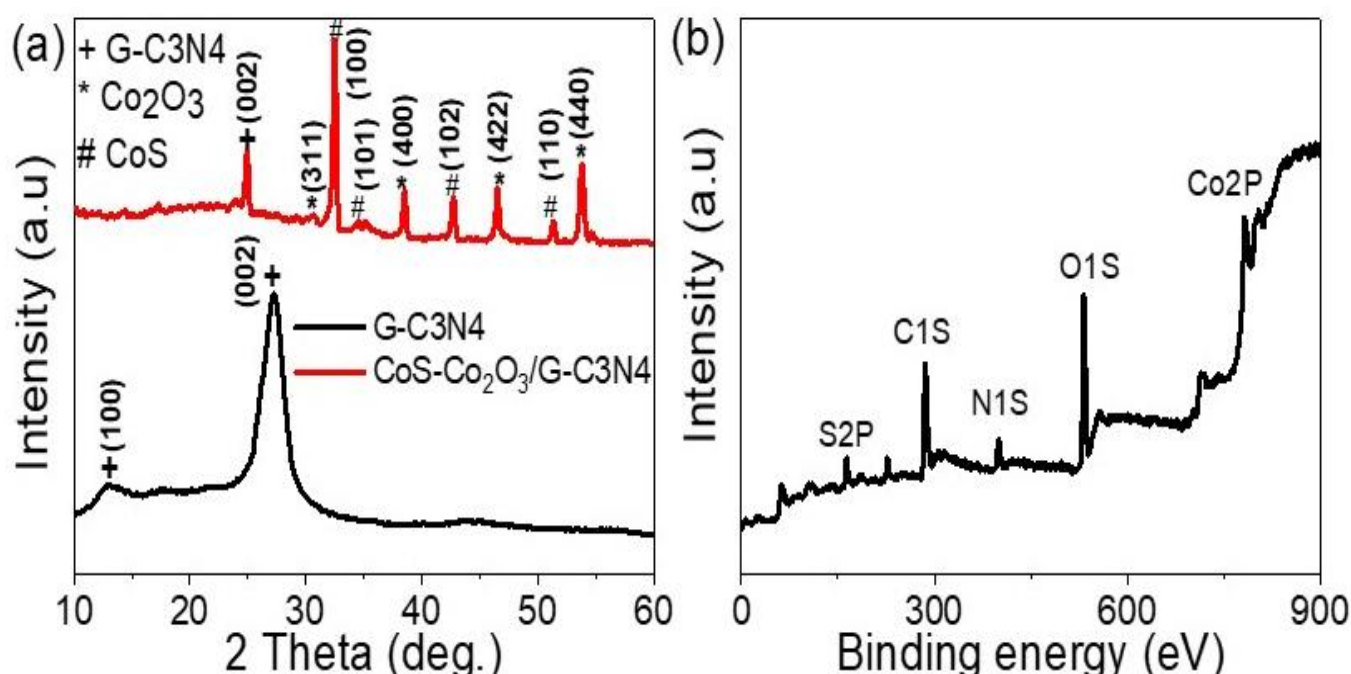


Figure 1. (a) XRD of G-C₃N₄ and CoS-Co₂O₃/G-C₃N₄ composite and (b) XPS of CoS-Co₂O₃/G-C₃N₄ composite.

The chemical and structural composition was investigated via X-ray photoelectron spectroscopy (XPS) (see Figure 1b). Numerous elements were located in the sample, such as O, C, Co, N, and S (see Table 1). The O element was located at 532.5 eV and resulted

from Co_2O_3 or oxygen inside the G-C3N4. Although the C1s spectrum was located at about 285.9 eV which can from the utilized pure G-C3N4, several bonds such as C-C, C-NH₂, and N-C=N bonding at about 284, 286, and 288, respectively, were also distinguished. A small shift of the C-C XPS peak to higher binding energy was located, representing the chemical bonding between G-C3N4 and CoS-Co₂O₃. The N 1s strong peak centering at 399.7 eV was due to the nitrogen atom inside the G-C3N4. The S2p peak was located at 163.1 eV, which could be assigned to S-Co bonding. Furthermore, a Co2p spectrum peak was located at 782.25 eV, which could be assigned to the high binding between CoS-Co₂O₃ and G-C3N4.

Table 1. The binding energy and percent of elements in the composite CoS-Co₂O₃/G-C3N4 were calculated from the XPS analysis.

Element	Peak (eV)	Atomic%
O1s	532.5	25.01
C1s	285.9	52.39
N1s	399.7	8.1
S2p	163.1	7.28
Co2p	782.2	7.22

The morphological and structural analysis of the samples were based on the correlation of SEM, TEM, and theoretical modeling images. Figure 2 represents SEM images of the (a) G-C3N4 and (b) CoS-Co₂O₃/G-C3N4 composites. The G-C3N4 has 2D nanosheet properties; the formed sheets enfold and crumple in some areas. The 2D nanosheets have an average width and length of 80 nm and 170 nm. It can be seen that both images reveal a thin sheet structure. Indeed, this was clearly distinguished through the TEM image (Figure 2c) of the G-C3N4, illustrating flat irregular shaped sheets with wrinkles. The dark area covering the sheets confirms CoS-Co₂O₃ incorporation in G-C3N4 sheets.

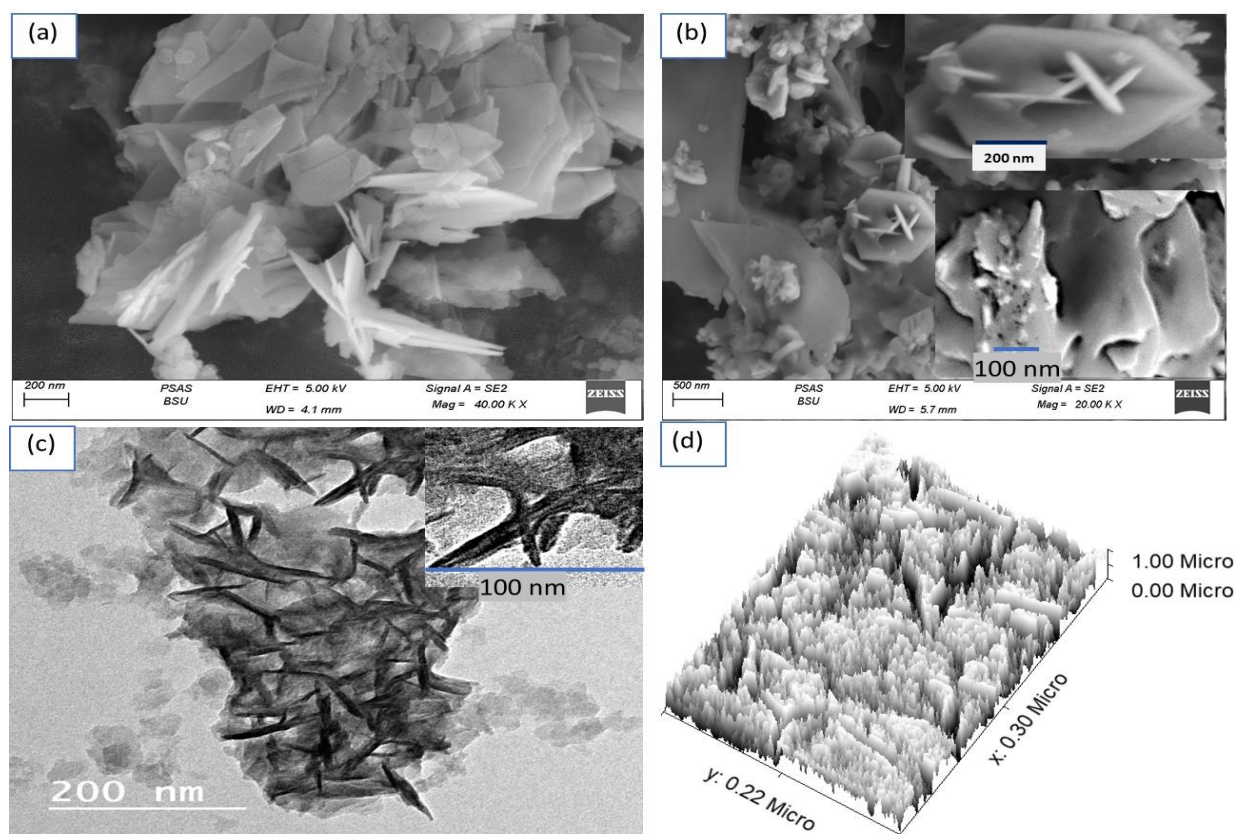


Figure 2. SEM of (a) G-C3N4 and (b) CoS-Co₂O₃/G-C3N4 composite. (c) TEM and (d) theoretical image of CoS-Co₂O₃/G-C3N4 composite.

The theoretical modeling is shown in Figure 2d; the roughness and cross-section are clearly displayed. The formed CoS-Co₂O₃/G C3N4 composite is highly homogeneous roughness, in which the small particles coat the sheets represented by the G-C3N4.

The Electrochemical Study of the Prepared Supercapacitor

The charge and discharge behavior of CoS-Co₂O₃/G-C3N4 composite nanomaterials were studied under different electrolytes; 6 M NaOH and 0.5 M HCL (see Figure 3a,b). Measurements were carried out in a potential window (0 to 1.0 V) at 25 °C, through a current density of 0.4 to 1.0 A/g. Moreover, the supercapacitor based on two symmetric structured electrodes was used, in which 0.0003 g paste was loaded over each electrode; then, a filter paper saturated with the selected electrolyte was used as a separator between these two electrodes. From the curve behavior, the capacitive properties affected the current density, which decreased with an increase in the inserted current density. Furthermore, the capacitive properties had a greater value in the basic medium in comparison with the acid medium. This is related to the effect of the HCl electrolyte on Co₂O₃ under the reaction of an acid with a base. The charge and discharge lifetime values in the acid and base mediums were 85 and 456 s, respectively. This confirmed the non-linear curves and change in behavior in the base medium [22–24]. The formation of the OH[−] group caused increasing negative charge on the supercapacitor electrode that enhanced the pseudocapacitance properties of the prepared supercapacitor. On the other hand, the H⁺ ions caused the same effect, but the acid-base reaction under the presence of Co₂O₃ decreased the total capacitance.

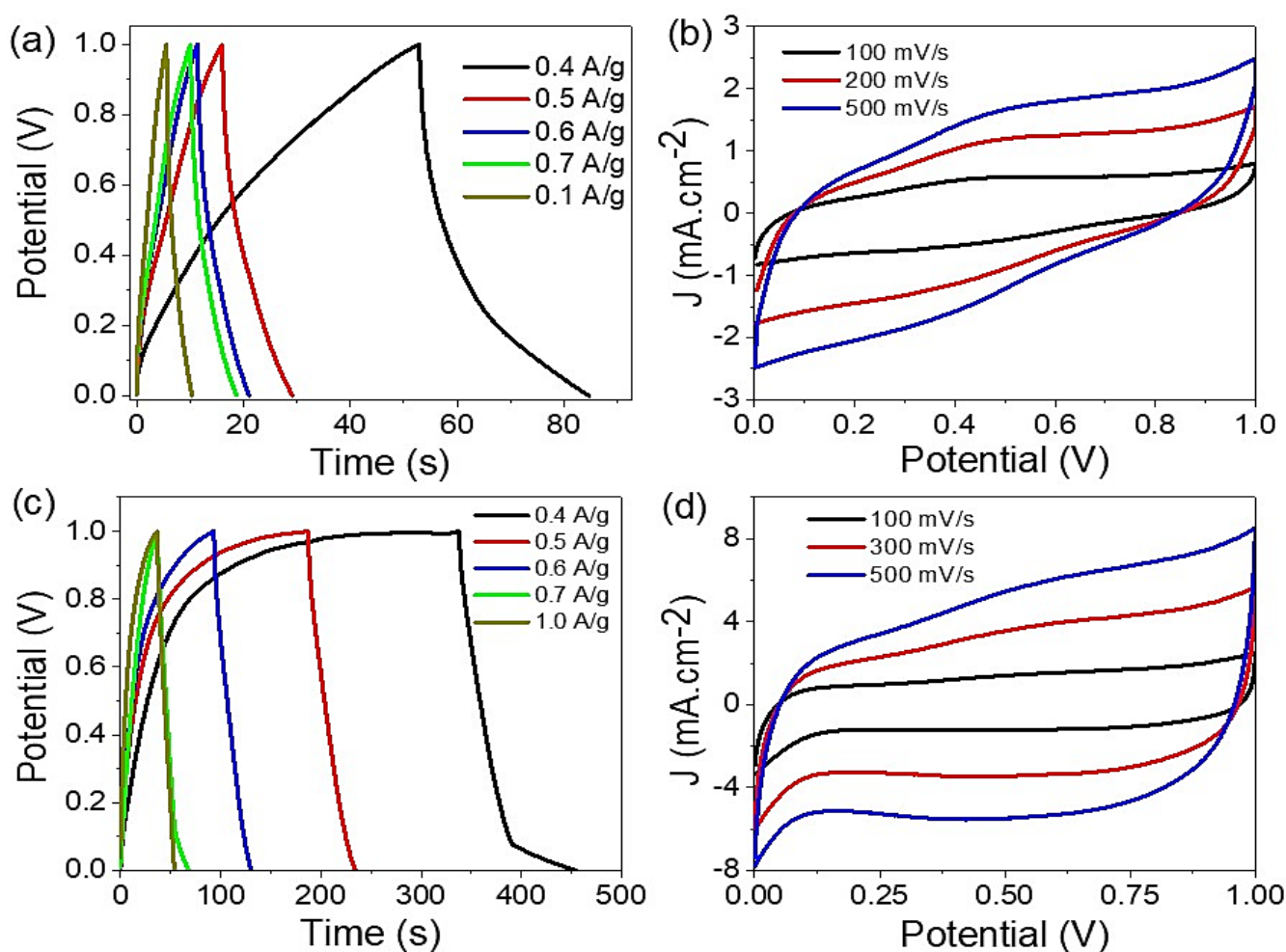


Figure 3. (a,c) Charge and discharge and (b,d) cyclic voltammetry curves for CoS-Co₂O₃/G-C3N4 nanocomposite supercapacitor under HCl and NaOH electrolytes, respectively.

The cyclic voltammetry behavior confirmed the charge and discharge behavior [25,26], where the produced current density values had a greater value in the base medium than in the acid medium (see Figure 3c,d). The rectangular shape appeared in the base medium, which confirmed the pseudo-capacitance feature [27,28], in which the surface area increased by increasing the scan rate from 100 to 500 mV.s^{-1} . This confirmed the diffusion time for the electrolyte to diffuse inside the supercapacitor paste. The oxidation and reduction behavior increased on both sides of the cyclic voltammetry curve under the rectangular formed shape. The cyclic voltammetry behavior depends on the electrolyte used, in which the rectangular shape is confirmed from a base medium for the pseudo-capacitance feature.

The stability of the supercapacitor electrodes is determined through the charge and discharge behavior at a current density of 0.4 A/g for 1000 cycles using two different electrolytes, 0.5 M HCl and 6 M NaOH (see, Figure 4a,b). From these figures, the two supercapacitors in both the acid and base mediums have greater stability up to 1000 cycles, but the supercapacitor in the base medium obtains better behavior. Figure 4c gives the relation of cycle numbers and the capacitance retention in both acid and base mediums. From this figure, both supercapacitors have great stability up to 400 cycles, but then the supercapacitor in the acid medium faces a small stability loss, which may be due to the reaction of HCl with the Co_2O_3 inside the paste of the electrode. The supercapacitor in the base medium has 100% stability retention up to 600 cycles; then, the stability decreases to 98.5% up to 1000 cycles.

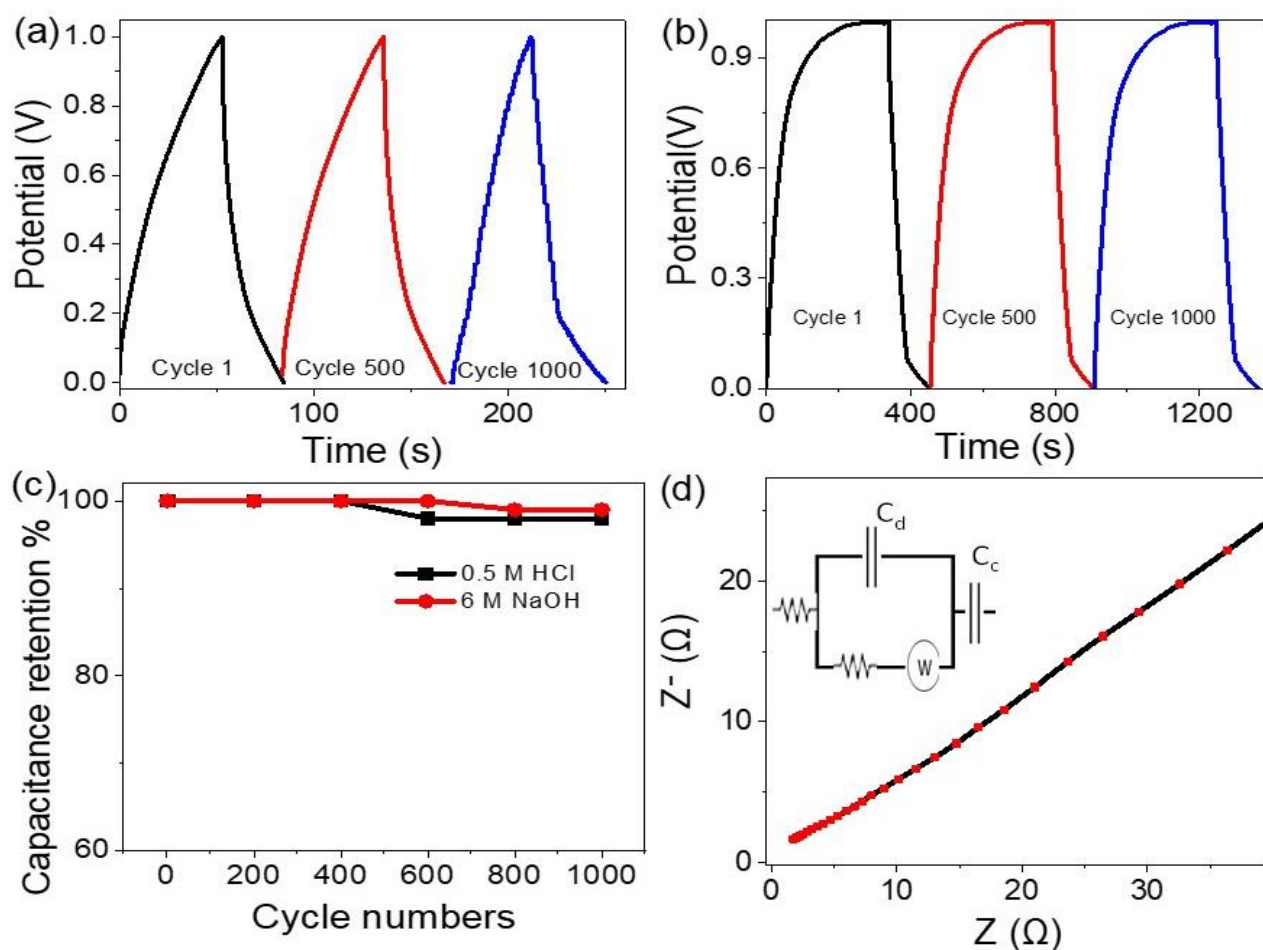


Figure 4. The cycle lifetime of the supercapacitor using the electrolytes (a) 0.5 M HCl and (b) M NaOH. (c) Capacitance retention using different electrolytes and (d) the impedance of the supercapacitor in basic medium 6 M NaOH.

The relation between the real (Z') and imaginary (Z'') impedance (Nyquist plot) is shown in Figure 4d for the supercapacitor in 6 M NaOH. Moreover, the impedance circuit represents the electrochemical contact of the prepared supercapacitor (see insert Figure 4d), in which the series and charge transfer resistance represent the electrolyte and electrolyte/electrode resistances, respectively, while the Warburg impedance represents the resistance of the electrode material [29,30]. Moreover, the capacitance appears in the circuit that represents the contact. The supercapacitor displays a very small Z'' value, confirming the greater behavior of NaOH as an electrolyte for charge transfer (R_{ct}) between the electrolyte and the electrode (see Figure 4d). R_{CT} characterizes the rate of redox reactions at the electrode/electrolyte interface, while C_{dl} occurs at the interfaces between solids and ionic solutions due to separations of ionic and electronic charges. The values of R_s and R_{CT} are 1.54 and 6.4 Ω , respectively, while the C_{dl} value is 18 μF .

For calculation of the performance of the prepared supercapacitor, the specific capacitance (C_s) was determined in the acid and base mediums (see, Figure 5a). Such a calculation is based on Equation (1) [31,32], in which I , Δt , ΔV , and m represent current (A), discharge time (s), potential window (V), and the paste mass (g), respectively. The supercapacitor has C_s values of 361 and 92 F/g in the basic and acidic mediums, respectively. In addition, the energy density (E) represents the capacitance of the prepared supercapacitor. These E values were calculated for the supercapacitor from 6 M NaOH and 0.5 M HCl, as shown in Figure 5b and Equation (2). This E value depends on potential windows V_{max} and V_{min} , in which the produced E values in the acid and base mediums are 28.7 and 30.2 $W h kg^{-1}$, respectively.

$$C_s = 4I\Delta t / \Delta V \cdot m \quad (1)$$

$$E = 0.5C_{sp} \cdot (V_{max}^2 - V_{min}^2) \quad (2)$$

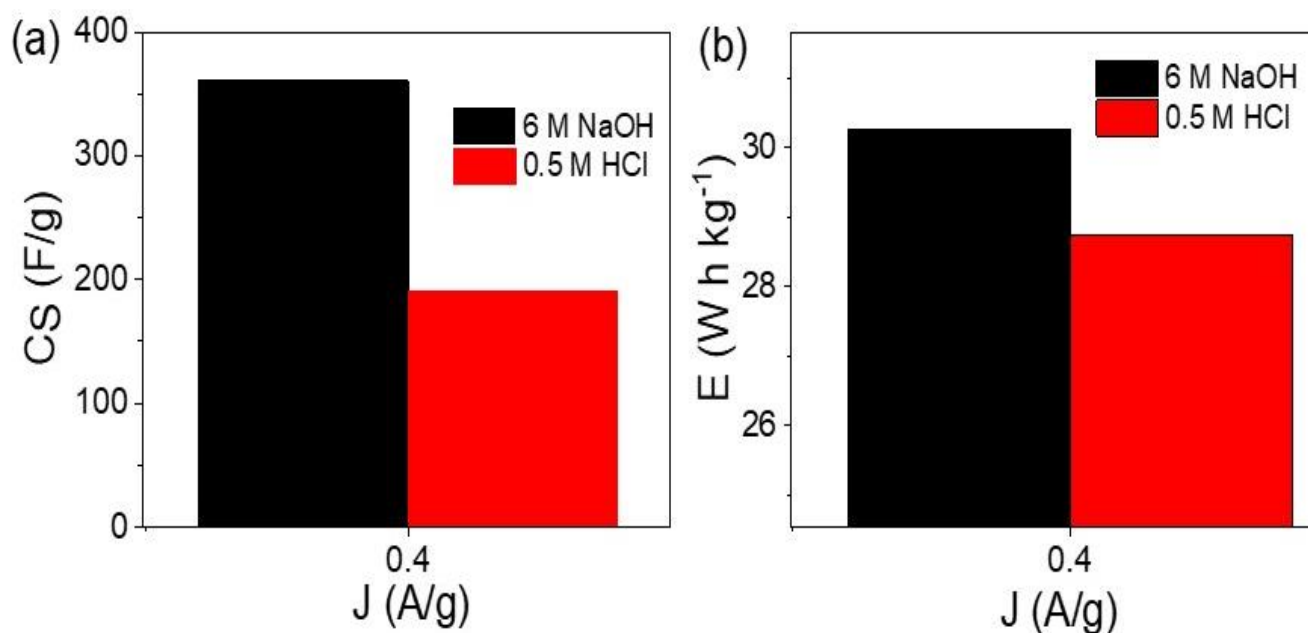


Figure 5. (a) The specific capacitance and (b) energy density for the prepared two symmetric electrodes supercapacitor in acid and base mediums.

4. Conclusions

Flower-shaped $CoS-Co_2O_3/G-C_3N_4$ composite was prepared using the hydrothermal method for two-symmetric-electrodes supercapacitor application. Such a composite was based on $G-C_3N_4$, first prepared using the combustion process in ambient air. The morphological properties were confirmed through SEM and TEM analyses. The chemical structures were confirmed using XRD and XPS analyses, in which all the functional groups

and elements were confirmed. The electrochemical studies were carried out by testing the prepared supercapacitor from the HCl and NaOH mediums. The supercapacitor parameters, charge/discharge, cyclic voltammetry, stability, and impedance of the prepared supercapacitor, were studied. The supercapacitor had a significant response in the basic medium in comparison to the acid medium. The supercapacitor had 100 and 98.5% stability retention up to 600 and 1000 cycles, respectively. The supercapacitor had C_s values of 361 and 92 F/g in the basic and acidic mediums, respectively. Moreover, the E values were 28.7 and 30.2 W h kg⁻¹, respectively. This promising prepared supercapacitor device has great electrochemical properties that qualifies it for industrial applications.

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