

**MINISTRY OF EDUCATION  
AND TRAINING**

**VIETNAM ACADEMY OF  
SCIENCE AND TECHNOLOGY**

**GRADUATE UNIVERSITY OF SCIENCE AND TECHNOLOGY**

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**LE HONG QUAN**

**PREPARATION OF CARBON AEROGEL FROM CHITOSAN  
AND THEIR COMBINATION WITH Ni, Co OXIDE/SULFIDE  
FOR USE AS ELECTRODES IN ASYMMETRIC  
SUPERCAPACITORS**

**SUMMARY OF DISSERTATION ON SCIENCES OF MATTER**

Major: Materials for Electronics

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The thesis is completed at Graduate University of Science and Technology, Vietnam Academy of Science and Technology.

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**PREFACE**

**The necessary of the thesis**

The fourth industrial revolution and industrial and population growth significantly impact many areas, including energy. Efforts are being made to promote renewable energy solutions, but many challenges exist in efficiently converting, storing, and distributing this energy source. Supercapacitors are devices with great potential for this purpose today. The ideal electrode material should have the following properties: large capacitance, high conductivity, large surface area, low density, large porosity with suitable pore size, and additional redox reactions. To achieve this, it is necessary to use composite materials of two or three components, including metal oxide/hydroxide/sulfide materials, carbon, and conductive polymers. Based on this reality, in this thesis, we choose the topic: *“Preparation of carbon aerogel from chitosan and their combination with Ni, Co oxide/sulfide for use as electrodes in asymmetric supercapacitors.”*

**The objectives of the thesis**

Successful fabrication of aerogel carbon from chitosan, composite containing Ni, Co oxide/sulfide, and carbon from chitosan with properties meeting the requirements for electrode materials for supercapacitors.

**The thesis contents:**

- Preparation and characterization of various aerogel carbon from chitosan, composites consisting of Ni, Co oxide/sulfide, and carbon from chitosan.
- Analyze the materials' properties and determine the electrodes' electrochemical parameters.
- Investigation of the electrochemical performance of the supercapacitor.

**The thesis layout:**

The thesis contains 134 pages, 106 figures, 17 tables, and 191 references. It includes the following parts: Introduction, five chapters, and Conclusion.

The results were published in 03 articles in SCIE journals and published in 01 article in an international conference proceeding.

## **CHAPTER 1. INTRODUCTION**

Chapter 1 has 21 pages, 11 figures, including the following sections:

### **1.1 Supercapacitors**

#### **1.1.1 Supercapacitor setup**

#### **1.1.2 Operating principle**

### **1.2 The electrode active materials**

Researchers and developers are aiming to research composite materials containing carbon and metal oxides/sulfides to take advantage of the benefits of each individual material.

#### **1.2.1 Carbon-based materials**

N-containing carbon aerogels derived from chitosan are currently the focus of research for creating electrodes with high specific capacitance.

#### **1.2.2 Metal oxide/sulfides**

#### **1.2.3 Carbon-based and metal oxides/sulfide composites**

Carbon-based materials with good electrical conductivity and high surface area can attach nano-sized metal oxides/sulfides, providing maximum electrical active area. Furthermore, metal oxides/sulfides can effectively promote charge transfer and enhance the electrochemical performance of supercapacitors through redox reactions.

### **1.3 Studies in Vietnam**

**Conclusion of Chapter 1:** After reviewing the literature, it is clear that there is a research gap in using seafood waste to produce aerogel composite materials. These materials would consist of Ni, Co oxide/sulfide, and carbon from chitosan and serve as electrode materials for supercapacitors. Most existing studies concentrate on individual materials, such as carbon from chitosan and metal oxide/sulfide. However, combining carbon from chitosan with Ni, Co oxide/sulfide could improve electrochemical performance.

Additionally, creating composite materials containing carbon from chitosan could support sustainable development in the aquaculture and seafood processing industry.

## CHAPTER 2. EXPERIMENTAL

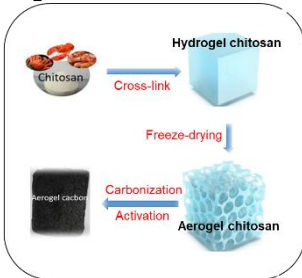
Chapter 2 has 23 pages, 23 figures, and 3 tables including the following sections:

### 2.1 Preparation of carbon aerogel from chitosan

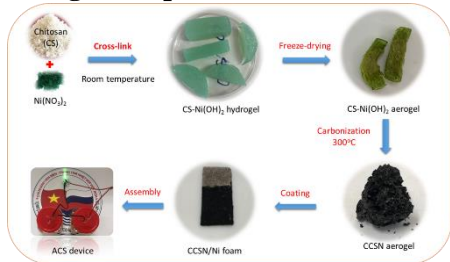
### 2.2 Preparation of NiO/carbon aerogel composites

### 2.3 Preparation of NiCo<sub>2</sub>O<sub>4</sub>/carbon aerogel composites

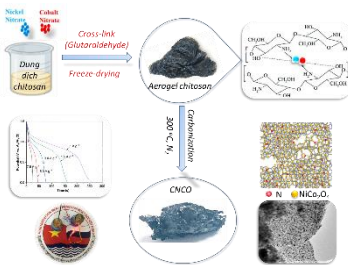
### 2.4 Preparation of NiCo<sub>2</sub>S<sub>4</sub>/carbon aerogel composites



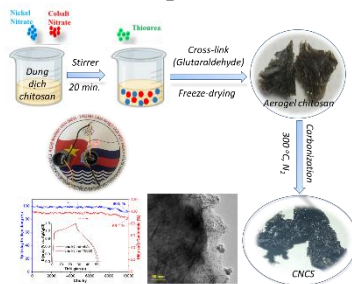
**Fig. 2.2.** Preparation scheme of carbon aerogel from chitosan



**Fig. 2.5.** Preparation scheme of NiO/carbon from chitosan composites



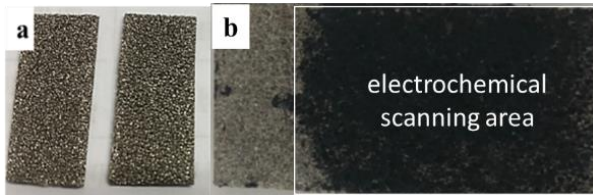
**Fig. 2.6.** Preparation scheme of NiCo<sub>2</sub>O<sub>4</sub>/carbon from chitosan composites



**Fig 2.7.** Preparation scheme of NiCo<sub>2</sub>S<sub>4</sub>/carbon from chitosan composites

## 2.5 Fabrication of supercapacitor electrodes

The electrode materials, including active material, carbon black powder, and binder, were taken in a mass ratio of 8:1:1. The mixture was stirred using ultrasonic waves until wholly mixed, specifically at 250 kHz for 30 seconds. Next, apply the mix mentioned above onto the nickel foam using the drop-casting method.



**Fig. 2.8.** (a) Nickel foam sheet, (b) Supercapacitor electrode

## 2.6 Fabrication of supercapacitor devices

The supercapacitor device was asymmetric, with composite material as the positive electrode and aerogel carbon from chitosan as the negative electrode. Solid electrolyte from KOH and PVA. The separator was glass fiber filter paper.

## 2.7 Methods for studying the micromorphology and structure of materials

### 2.7.1 Scanning electron microscope

### 2.7.2 Transmission electron microscope

### 2.7.3 X-ray diffraction

### 2.7.4 Fourier-transform infrared spectroscopy

### 2.7.5 Nitrogen adsorption-desorption isotherm

## 2.8 Electrochemical performance tests

### 2.8.1 Cyclic voltammetry (CV)

### 2.8.2 Galvanostatic charge-discharge (GCD)

### 2.8.3 Electrochemical impedance measurement (EIS)

### **2.8.4 Determination of electrochemical parameters**

The electrochemical properties of materials and supercapacitors were measured using the Autolab PGSTAT302N electrochemical measuring instrument, which included specific capacitance, power density, energy density, and cycle durability.

### **2.8.5 Determine the capacitive contribution of material component**

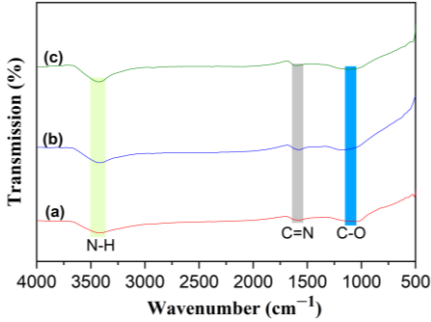
**Conclusion of Chapter 2:** This chapter has outlined the experimental methods utilized in the thesis, including:

- i. The sol-gel method utilizes glutaraldehyde crosslinker to form a chitosan hydrogel, which is then freeze-dried to obtain chitosan aerogel, and finally carbonized in an N<sub>2</sub> gas environment. This method enables the fabrication of large-scale, uniform, and highly stable samples.
- ii. The drop-casting method was used to fabricate the electrode. Specifically, the electrode material solution is directly dropped onto the surface of the nickel foam. This method allows control of the electrode material mass.
- iii. The asymmetric supercapacitor is constructed with the following components: solid electrolyte, glass fiber insulator, negative electrodes using carbon aerogel material from chitosan, positive electrodes using composite material containing Ni, Co oxide/sulfide, and carbon from chitosan.
- iv. Methods for studying material micromorphology and structure: Scanning electron microscopy, transmission electron microscopy, X-ray diffraction, Fourier transform infrared spectroscopy, and nitrogen adsorption-desorption isotherms.
- v. Electrochemical parameters of electrodes and supercapacitors were determined using measurement methods such as cyclic voltammetry, galvanostatic charge-discharge, and electrochemical impedance measurement.

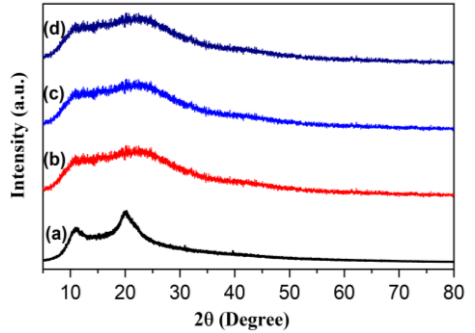
## CHAPTER 3. RESULTS OF MATERIAL PREPARATION RESEARCH

Chapter 3 has 22 pages, 27 figures, and 8 tables, including the following sections:

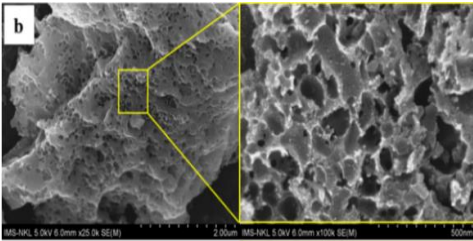
### 3.1 Aerogel carbon from chitosan



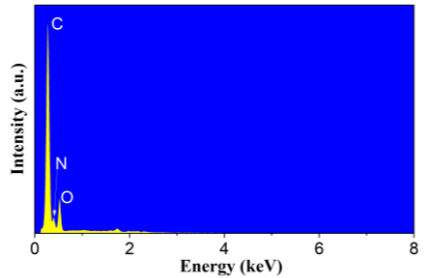
**Fig. 3.4.** Spectra FTIR: (a) ACCS-1, (b) ACCS-2, (c) ACCS-3



**Fig. 3.5.** Patterns XRD: (a) powder CS, (b) ACCS-1, (c) ACCS-2, (d) ACCS-3



**Fig. 3.7.** SEM images (a) ACCS-1, (b) ACCS-2 and (c) ACCS-3



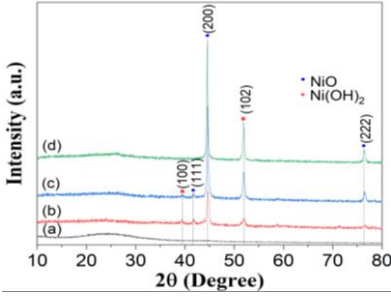
**Fig. 3.8.** Spectra EDS of ACCS-2

**Tab. 3.1.** Parameters of specific surface area, pore volume, and pore diameter of ACCS samples

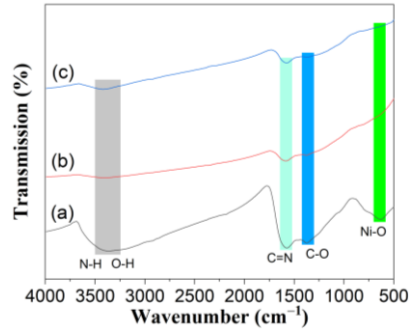
Mẫu	$S_{BET}$ ( $m^2 \cdot g^{-1}$ )	$V_{BJH}$ ( $cm^3 \cdot g^{-1}$ )	$d_{BJH}$ (nm)
ACCS-1	1533	0.706	0.97
ACCS-2	2341	1.29	0.99
ACCS-3	1714	0.98	1.04



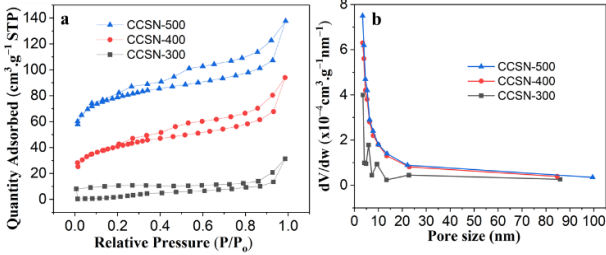
### 3.2 The NiO/carbon aerogel composites



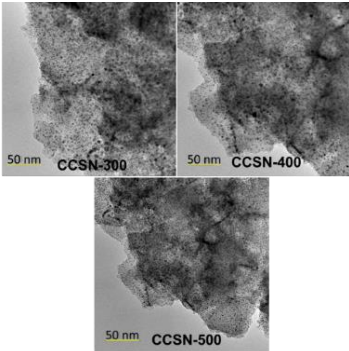
**Fig. 3.9.** Patterns XRD of (a) CCS, (b) CCSN-500, (c) CCSN-400 and (d) CCSN-300



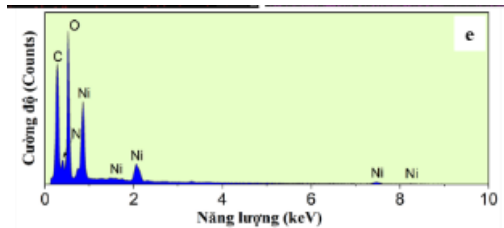
**Fig. 3.10.** Spectra FTIR of (a) CNCS-300, (b) CNCS-400 and (c) CNCS-500



**Fig. 3.11.** (a) BET and (b) pore size distribution of CCSN samples

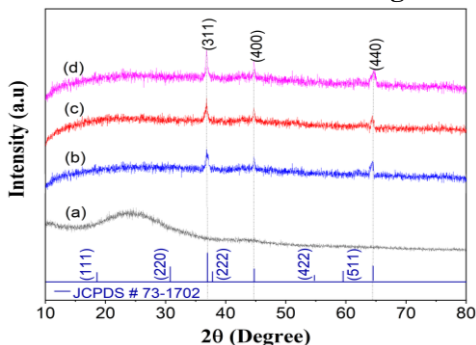


**Fig. 3.13.** TEM images of CCSN

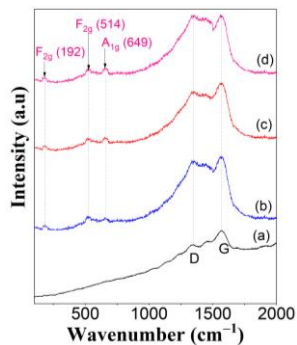


**Fig. 3.14.** (e) Spectra EDX of CCSN-300

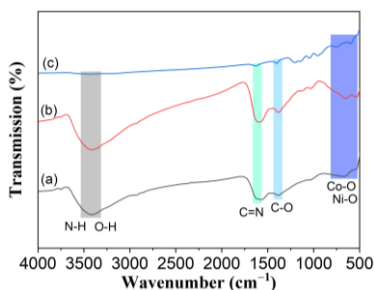
### 3.3 The NiCo<sub>2</sub>O<sub>4</sub>/carbon aerogel composites



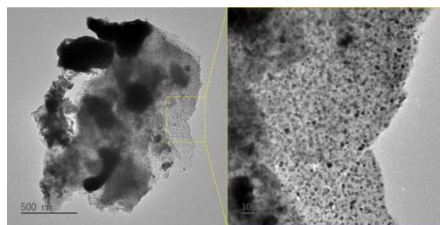
**Fig. 3.15.** Patterns XRD of (a) CCS, (b) CNCO-1, (c) CNCO-2 and (d) CNCO-3



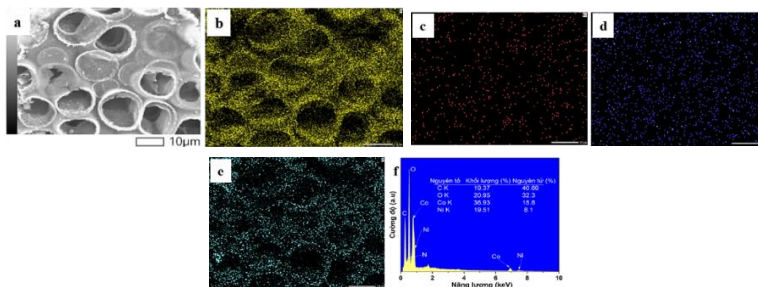
**Fig. 3.16.** Spectra Raman of (a) CCS, (b) CNCO-1, (c) CNCO-2 and (d) CNCO-3



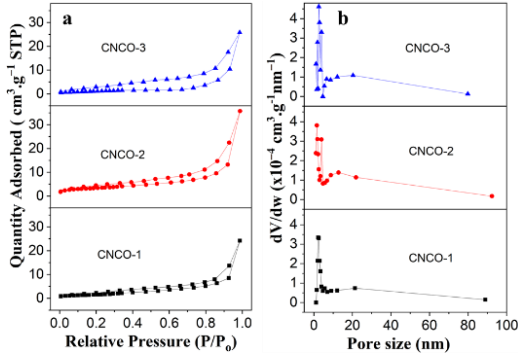
**Fig. 3.17.** Spectra FTIR of (a) CNCO-1, (b) CNCO-2 and (c) CNCO-3



**Fig. 3.19.** TEM image of CNCO-2

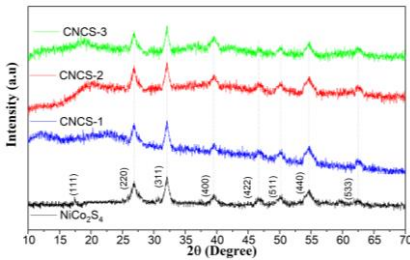


**Fig. 3.20.** (a) SEM image of CNCO-2, (b-e) EDS mapping of C, Ni, Co and O of the CNCO-2, f) EDS of CNCO-2

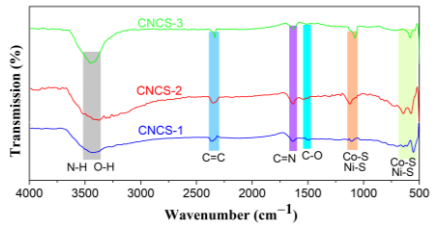


**Fig. 3.21.** (a) BET and (b) pore size distribution of CNCO sample

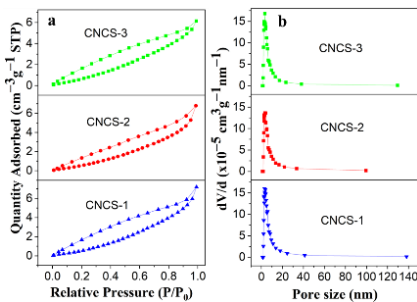
### 3.4 The NiCo<sub>2</sub>S<sub>4</sub>/carbon aerogel composites



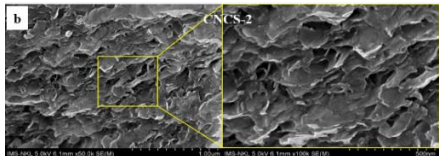
**Fig. 3.22.** Patterns XRD of NiCo<sub>2</sub>S<sub>4</sub> và CNCS



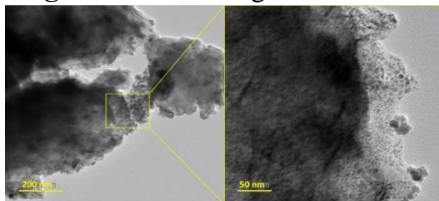
**Fig. 3.23.** Spectra FTIR of CNCS



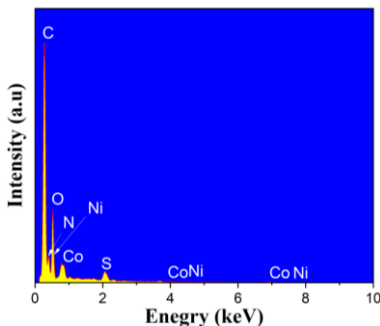
**Fig. 3.24.** (a) BET and (b) pore size distribution of CNCS samples



**Fig. 3.25.** SEM image of CNCS-2



**Fig. 3.26.** TEM image of CNCS-2



**Fig. 3.27.** (f) Spectra EDX of CNCS-2

**Conclusion of Chapter 3.** This chapter has provided an overview of material manufacturing processes and their corresponding material properties. Specifically:

- i. The carbon aerogel material was prepared from chitosan using the steps: gelation, freeze-drying, and carbonization. Glutaraldehyde was used as a cross-linking agent. The results showed that when using 2.5 ml of GA (1 wt.%) in 100 ml of chitosan solution (2.5 wt.%), the obtained carbon aerogel material had high porosity with a specific surface area of  $2341 \text{ m}^2 \cdot \text{g}^{-1}$ , pores distributed in various diameters (with an average of about 0.99 nm), and nitrogen content ranging from 8.1 to 9.3 (atom %).
- ii. The following steps were taken to prepare composite materials containing NiO and carbon from chitosan: gel formation, freeze-drying, and carbonization. The best ratio chosen was 6 mmol of  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  precursor in 100 ml chitosan solution (2.5 wt.%), with a calcination temperature of  $300^\circ\text{C}$ . The resulting composite materials exhibited a uniform distribution of NiO nanoparticles on a carbon matrix derived from chitosan. The nitrogen content was approximately 4.5 to 5.2 (atom %), the average pore diameter was about 3.4 nm, and the average size of NiO nanoparticles was approximately 16 to 20 nm.
- iii. Composite materials containing  $\text{NiCo}_2\text{O}_4$  and carbon from chitosan were prepared through gelation, freeze-drying, and carbonization. The chosen

precursor ratio was approximately 2 mmol of  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and 4 mmol of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  in 100 ml chitosan solution (2.5 wt.%), and the calcination temperature was  $300^\circ\text{C}$ . The resulting composite materials exhibited a uniform distribution of  $\text{NiCo}_2\text{O}_4$  nanoparticles on the carbon matrix from chitosan, with a nitrogen content of about 2.9 to 4.7 (atom %). The average size of the  $\text{NiCo}_2\text{O}_4$  nanoparticles was approximately 14 to 21 nm.

iv. Composite materials containing  $\text{NiCo}_2\text{S}_4$  and carbon from chitosan were created through a series of steps including gelation, freeze-drying, and carbonization. The chosen precursor ratio was approximately 2 mmol  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , 4 mmol  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , and 8 mmol  $\text{CH}_4\text{N}_2\text{S}$  in 100 ml chitosan solution (2.5 wt.%), with a calcination temperature of  $300^\circ\text{C}$ . The resulting composite materials featured a uniform distribution of  $\text{NiCo}_2\text{S}_4$  nanoparticles on the carbon matrix from chitosan, with a nitrogen content of about 10.4 to 11.7% (atom %). The average size of the  $\text{NiCo}_2\text{S}_4$  nanoparticles was approximately 8 to 15 nm.

## CHAPTER 4. RESULTS OF ELECTRODE FABRICATION RESEARCH

Chapter 4 has 20 pages, 25 figures, and 3 tables, including the following sections:

### 4.1 Electrode using carbon aerogel material from chitosan

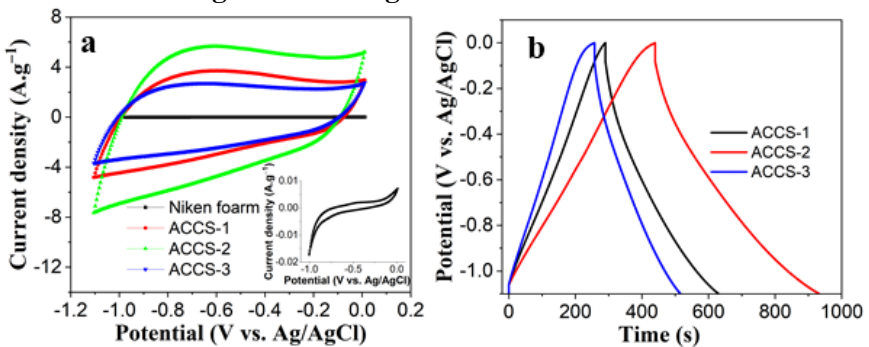
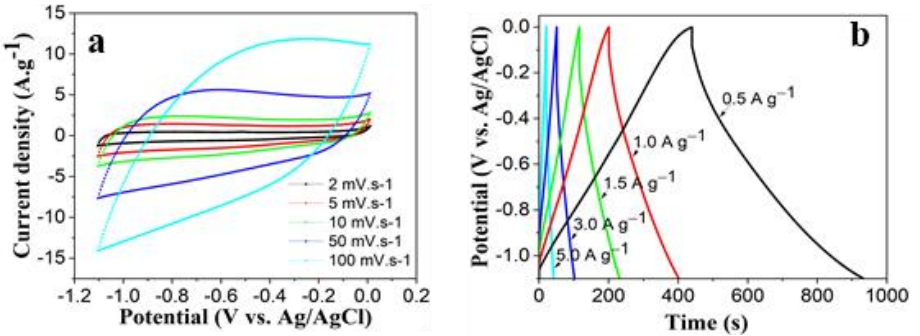
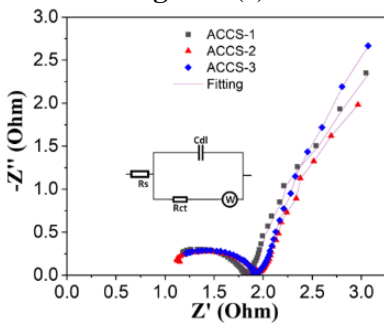


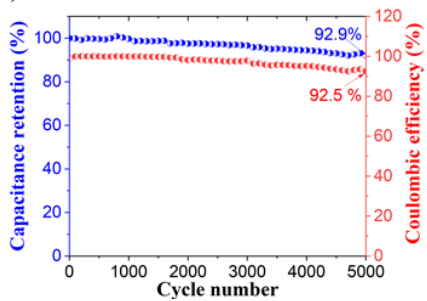
Fig. 4.1. (a) CV curves, (b) GCD curves



**Fig. 4.3.** (a) CV curves, (b) GCD curves of ACCS-2

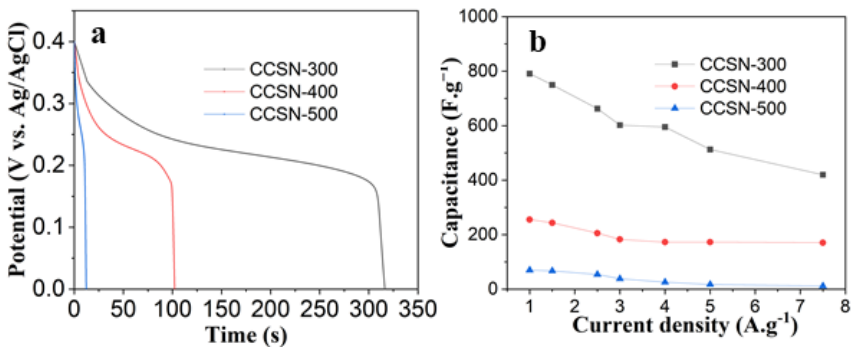


**Fig. 4.4.** EIS spectra



**Fig. 4.5.** The capacitance retention of ACCS-2 at 5 A.g<sup>-1</sup>

## 4.2 Electrode using NiO/carbon aerogel composites



**Fig. 4.8.** (a) GCD curves at 1 A.g<sup>-1</sup>, (b) Specific capacitance of CCSN at the different current density

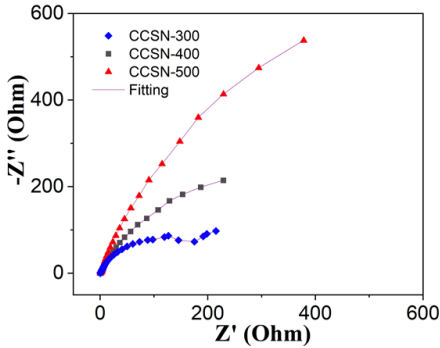


Fig. 4.9. EIS spectra

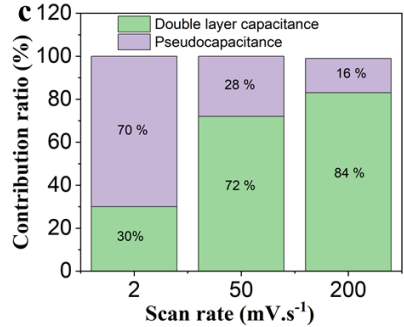


Fig. 4.12. (c) Capacitance contribution ratio

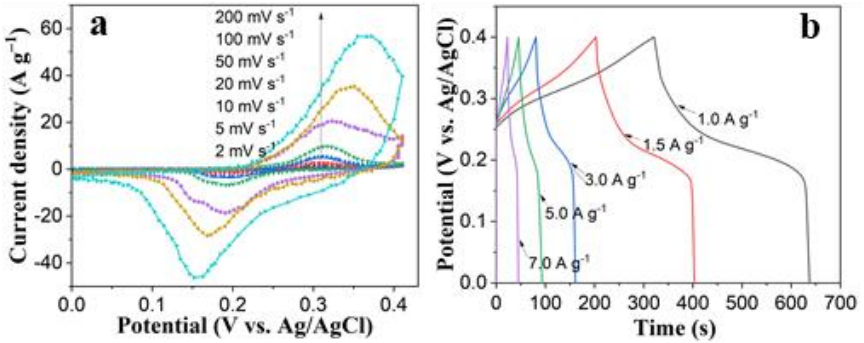
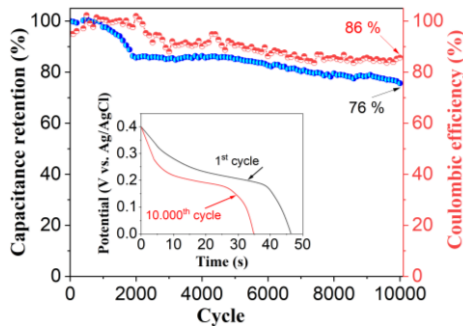
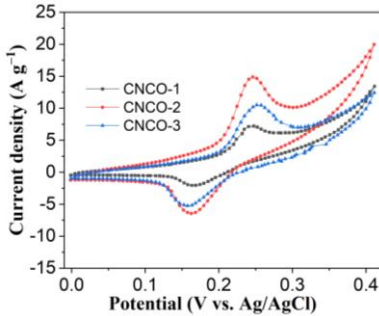


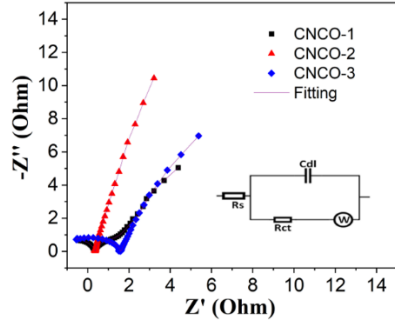
Fig. 4.11. (a) CV curves, (b) GCD curves of CCSN-2

Fig. 4.13. The capacitance retention of CCSN-2 at  $5.0\text{ A}\cdot\text{g}^{-1}$

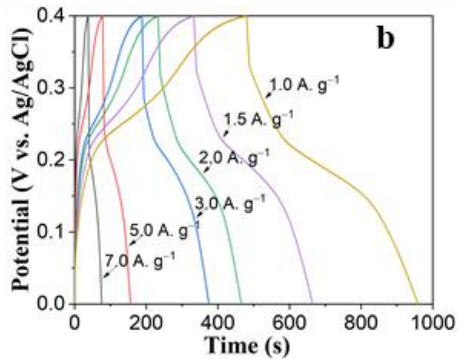
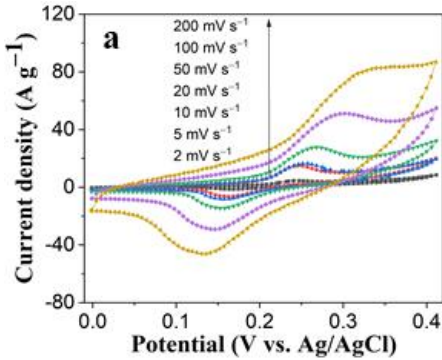
### 4.3 Electrode using $\text{NiCo}_2\text{O}_4$ /carbon aerogel composites



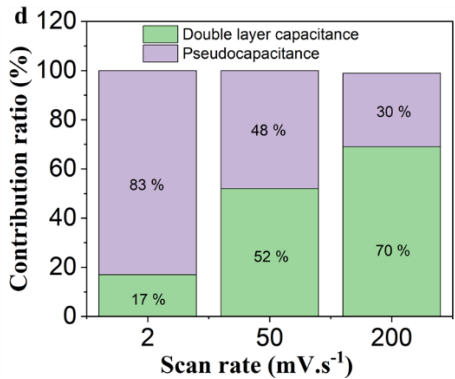
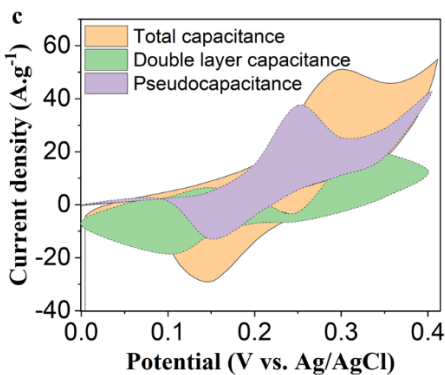
**Fig. 4.14.** CV curves of CNCO at 5  $\text{A.g}^{-1}$



**Fig. 4.15.** EIS spectra

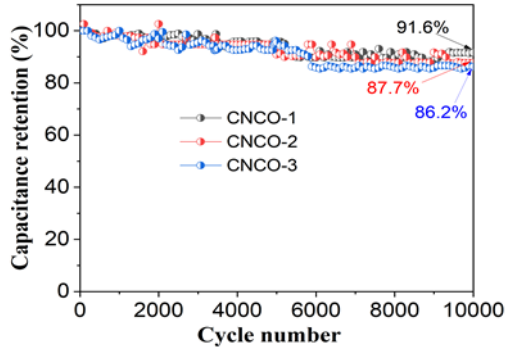


**Fig. 4.17.** (a) CV curves, (b) GCD curves of CNCS-2



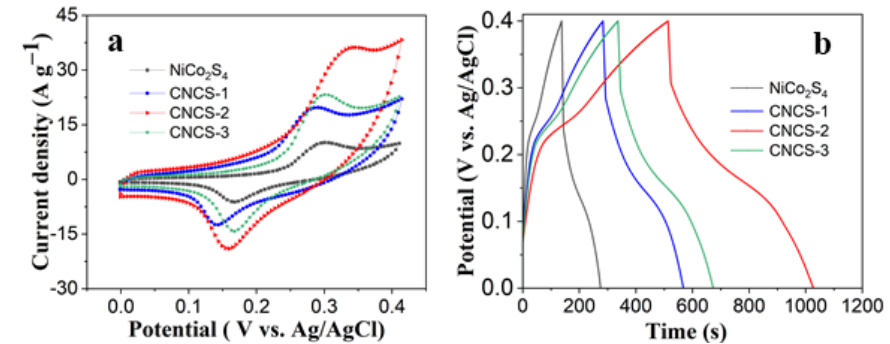
**Fig. 4.18.** (c) Component capacitance and (d) Capacitance contribution ratio



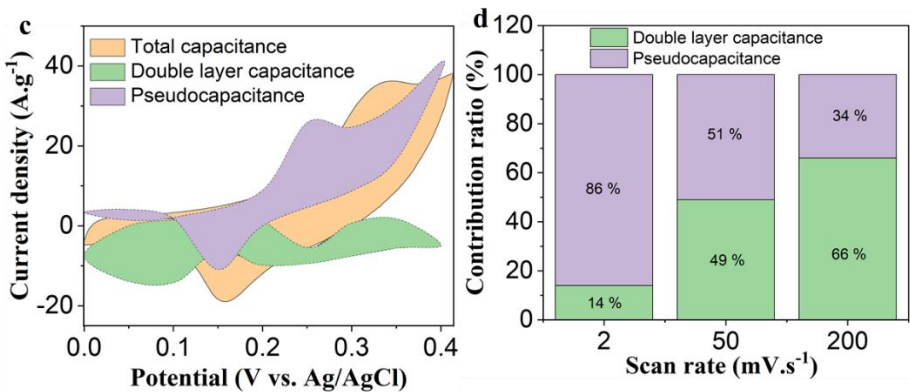


**Fig. 4.19.** The capacitance retention of CNCO at  $7 \text{ A.g}^{-1}$

#### 4.4 Electrode using $\text{NiCo}_2\text{S}_4$ /carbon aerogel composites



**Fig. 4.20.** (a) CV curves, (b) GCD curves.



**Fig. 4.21.** (c) Component capacitance and (d) Capacitance contribution ratio

**Conclusion of Chapter 4.** Electrodes have been created using carbon derived from chitosan and composite materials. Specifically:

i. The chitosan carbon aerogel, utilizing glutaraldehyde as a cross-linking agent at a ratio of 2.5 ml GA (1 wt%) in 100 ml of a 2.5 wt% chitosan solution, demonstrated the best electrochemical performance. It achieved a specific capacitance of  $183 \text{ F.g}^{-1}$  at a current density of  $1.0 \text{ A.g}^{-1}$ . Additionally, the capacitance retention rate exceeded 92 % after 5000 charge-discharge cycles at a current density of  $5 \text{ A.g}^{-1}$ .

ii. The ratio of nickel (Ni) to cobalt (Co) oxide/sulfide precursors significantly influences the electrochemical performance. A composite material composed of nickel oxide (NiO) and carbon derived from chitosan exhibited the best electrochemical properties when the precursor mass ratio was set to 6 mmol of  $(\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O})$  in 100 mL of a 2.5 wt% chitosan solution. Under these conditions, the specific capacitance achieved was  $790 \text{ F.g}^{-1}$  at a current density of  $1.0 \text{ A.g}^{-1}$ . However, the capacitance retention rate was low, reaching only about 76 % after 10000 charge-discharge cycles.

iii. The composite material containing  $\text{NiCo}_2\text{O}_4$  and carbon derived from chitosan demonstrated the best electrochemical properties when the precursor mass ratio was 2 mmol of  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and 4 mmol of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  in a 100 ml solution of chitosan (2.5 wt%). Specifically, the specific capacitance reached  $1200 \text{ F.g}^{-1}$  at a current density of  $1.0 \text{ A.g}^{-1}$ . It retained 86.2% of its capacitance after 10000 charge-discharge cycles, which was higher than that of the CCSN composite material.

iv. The composite material containing  $\text{NiCo}_2\text{S}_4$  and carbon derived from chitosan exhibited the most favourable electrochemical properties when prepared with a precursor mass ratio of 2 mmol  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , 4 mmol  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , and 8 mmol  $\text{CH}_4\text{N}_2\text{S}$  in 100 ml of a 2.5 wt% chitosan solution. The specific capacitance achieved was  $1282 \text{ F.g}^{-1}$  at a current density of  $1.0 \text{ A.g}^{-1}$ . It also retained 90.6 % of its capacitance after 10000

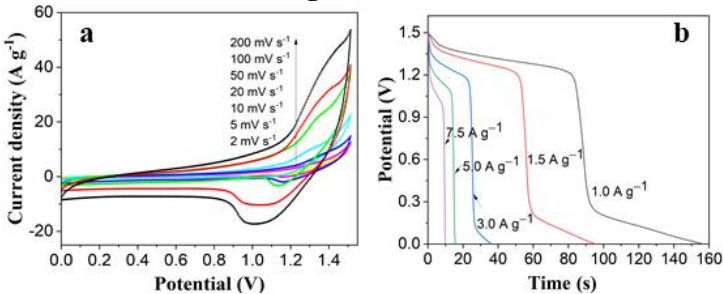
charge-discharge cycles, which was higher than that of the CCSN and CNCO composite electrodes.

v. The total capacitance of the composite material consists of two components: double-layer capacitance and pseudo-capacitance. The ratio of these components varies with the potential scan rate. As the scan rate increases, the contribution from double-layer capacitance rises, while the contribution from pseudo-capacitance decreases.

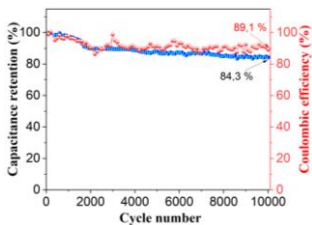
## CHAPTER 5. RESULTS OF SUPERCAPACITOR FABRICATION AND SURVEY

Chapter 5 has 14 pages, 20 figures, and 3 tables including the following sections:

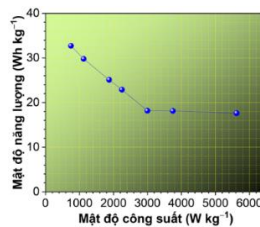
### 5.1 Asymmetric supercapacitor using a composite material containing NiO and carbon from chitosan as the positive electrode and carbon aerogel from chitosan as the negative electrode



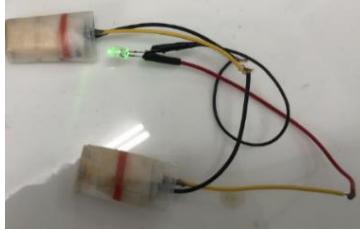
**Fig. 5.1.** (a) CV curves, (b) GCD curves of the ACCS/CCSN-2 device



**Fig. 5.3.** Capacitance retention and Coulombic efficiency of the ACCS/CCSN-2 device

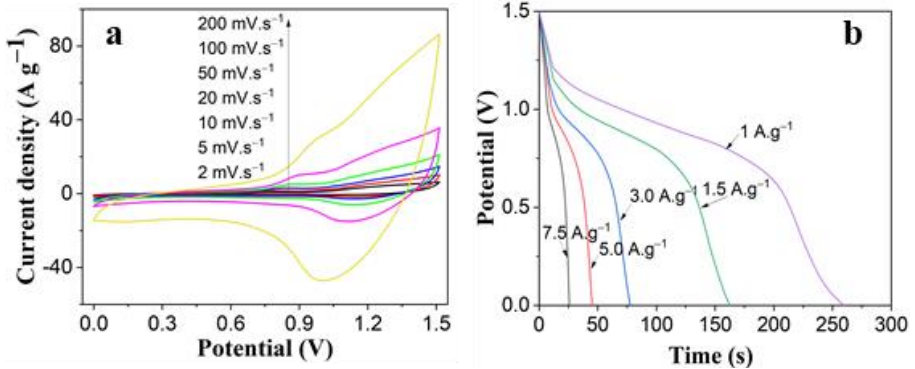


**Fig. 5.4.** Ragone diagram at different current densities of the ACCS/CCSN-2 device

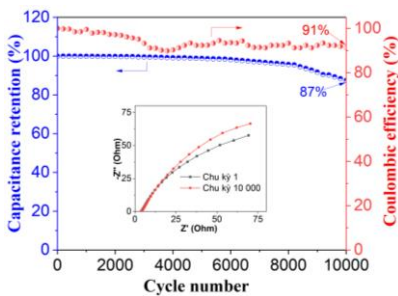


**Fig. 5.5.** Two ACCS/CCSN-2 devices are connected in series

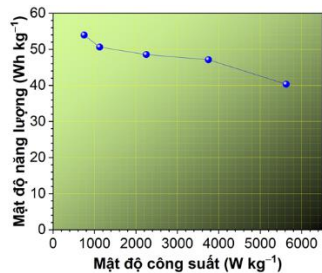
## 5.2 Asymmetric supercapacitor using a composite material containing $\text{NiCo}_2\text{O}_4$ and carbon from chitosan as the positive electrode and carbon aerogel from chitosan as the negative electrode



**Fig. 5.8.** (a) CV curves, (b) GCD curves of the ACCS/CNCO-2 device



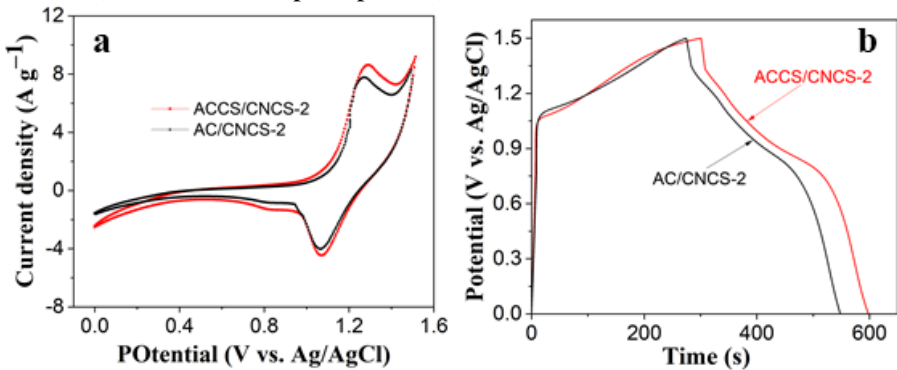
**Fig. 5.9.** Capacitance retention and Coulombic efficiency of the ACCS/CNCO-2 device



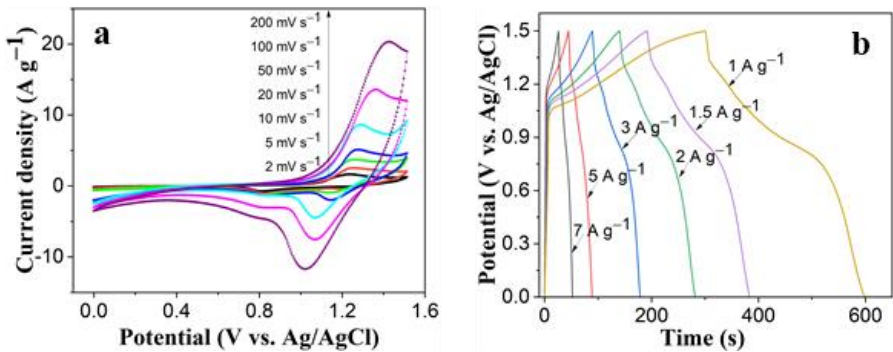
**Fig. 5.10.** Diagram at different current density of the ACCS/CNCO-2 device

### 5.3 Asymmetric supercapacitor using a composite material containing $\text{NiCo}_2\text{S}_4$ and carbon from chitosan as the positive electrode and carbon aerogel from chitosan as the negative electrode

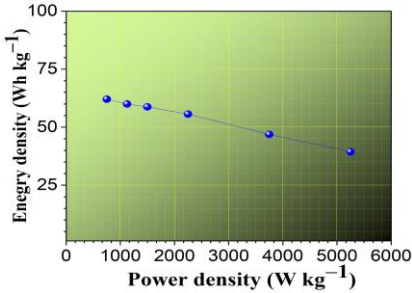
To further assess the effectiveness of using ACCS material as the negative electrode, we also utilized commercial activated carbon (AC) material as a control (AC/CNCS-2 Supercapacitor).



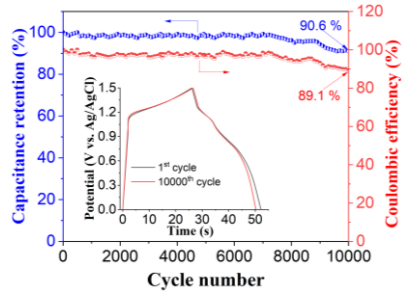
**Fig. 5.12.** (a) CV curves, (b) GCD curves of the ACCS/CNCS-2 and AC/CNCS-2 devices



**Fig. 5.13.** (a) CV curves, (b) GCD curves of the ACCS/CNCS-2 device

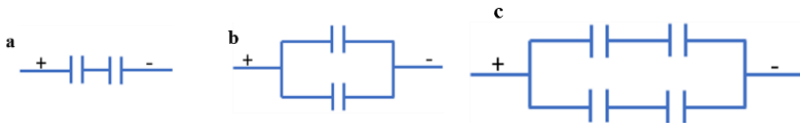


**Fig. 5.15.** Ragone diagram of ACCS/CNCS-2 device

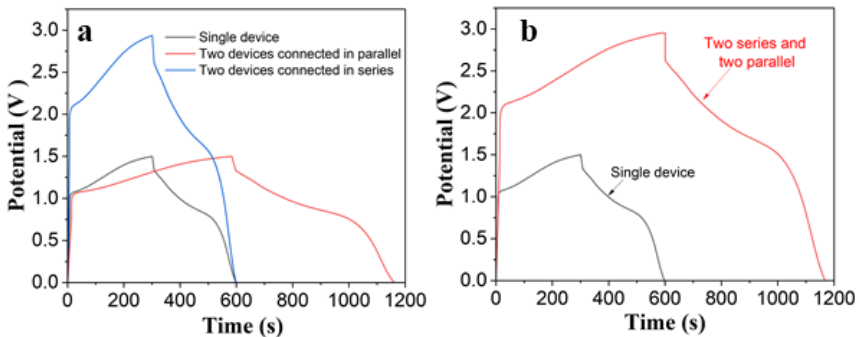


**Fig. 5.17.** Capacitance retention and Coulombic efficiency

The evaluation of the working system involves two individual supercapacitors connected in series (Fig. 5.18a), in parallel (Fig. 5.18b), and a two-stack system consisting of two series supercapacitors connected in parallel (Fig. 5.18c).



**Fig. 5.18.** (a) two devices connected in series, (b) two devices connected in parallel, (c) two clusters of two devices connected in series are connected in parallel



**Fig. 5.19.** (a) CV curves, (b) GCD curves



**Fig. 5.20.** Two ACCS/CNCS-2 devices are connected in series

**Conclusion of Chapter 5.** Asymmetric supercapacitors have been created using composite materials of Ni, Co oxide/sulfide, and carbon from chitosan as the positive electrode. The negative electrode is made from carbon aerogel derived from chitosan, and the solid electrolyte consists of KOH and PVA.

i. The maximum operating voltage of three supercapacitors, using chitosan carbon aerogel as the negative electrode and composite materials CCSN, CNCO, and CNCS as the positive electrodes, reached 1.5 V.

ii. The maximum energy density achieved was  $62 \text{ Wh.kg}^{-1}$  with the CNCS composite material as the positive electrode. Supercapacitors using the CNCO and CCSN composite materials demonstrated energy densities of  $53 \text{ Wh.kg}^{-1}$  and  $32 \text{ Wh.kg}^{-1}$ , respectively.

iii. The capacitance retention after 10000 charge/discharge cycles gradually decreased when using the composite materials in the order of CNCS, CNCO, and CCSN as positive electrodes, with corresponding values of 91%, 87%, and 84.3%, respectively.

iv. The supercapacitor using CNCS material as the positive electrode showed better electrochemical performance than CNCO and CCSN materials. Additionally, the supercapacitor employing carbon aerogel derived from chitosan as the negative electrode performed better than commercial activated carbon material because the carbon aerogel from chitosan has higher porosity, suitable pore distribution, and contains nitrogen in its structure.

## CONCLUSION

To achieve the goal of creating carbon aerogels from chitosan and composite materials containing Ni, Co oxides/sulfides, and carbon from chitosan with properties suitable for electrode materials for supercapacitors, the thesis focused on researching and fabricating these materials. Subsequently, asymmetric supercapacitors were developed using the researched materials, with a negative electrode utilizing carbon aerogels from chitosan and a positive electrode using composite materials containing Ni, Co oxides/sulfides, and carbon from chitosan. The research results lead to the following main conclusions:

1. Carbon aerogels and composites containing Ni, Co oxide/sulfide, and carbon were successfully prepared from chitosan. This was achieved through gelation steps using glutaraldehyde as a cross-linking agent, followed by freeze-drying and carbonization. The resulting materials exhibited high porosity, a large surface area, high available N content, and a uniform distribution of oxide/sulfide nanoparticles in the composite structure.
2. The electrodes, using composite materials containing (NiO, NiCo<sub>2</sub>O<sub>4</sub>, NiCo<sub>2</sub>S<sub>4</sub>) and carbon from chitosan, exhibited high specific capacitance values of 790, 1200, and 1282 F.g<sup>-1</sup> at a current density of 1.0 A.g<sup>-1</sup> in a 6M KOH electrolyte, respectively. The capacitance retention rates after 10000 charge/discharge cycles were 76 %, 87.7 %, and 90.6 %, respectively. Additionally, the electrode using carbon aerogel from chitosan demonstrated a specific capacitance of 183 F.g<sup>-1</sup> at a current density of 1.0 A.g<sup>-1</sup>, with a capacitance retention rate of over 92 % after 5000 charge/discharge cycles at a current density of 5 A.g<sup>-1</sup>.
3. The asymmetric supercapacitors utilized composite materials containing (NiO, NiCo<sub>2</sub>O<sub>4</sub>, and NiCo<sub>2</sub>S<sub>4</sub>) and carbon from chitosan as the positive electrode and carbon aerogel from chitosan as the negative electrode. They exhibited energy densities of 32, 52, and 62 Wh.kg<sup>-1</sup>, respectively, at a power



density of  $750 \text{ W.kg}^{-1}$ . After 10,000 charge/discharge cycles, the capacitance retention rates of the supercapacitors were 84.3 %, 87 %, and 89 %, respectively, and the Coulombic efficiencies were 89.1 %, 91 %, and 90.6 %.

The thesis has produced significant scientific findings. However, further research is required to use these materials as electrodes for supercapacitors. This includes studying the impact of nitrogen content on electrochemical performance, improving large-scale manufacturing capabilities, and reducing costs. We plan to continue investigating these areas in the future, hoping to achieve more compelling results and eventually move toward commercial production.

**LIST OF THE PUBLICATIONS RELATED TO THE  
DISSERTATION**

1. Le Hong Quan, Ung Thi Dieu Thuy, Pham Viet Nam, Nguyen Van Chi, Tang Xuan Duong, Nguyen Van Hoa, Chitosan-derived carbon aerogel nanocomposite as an active electrode material for high-performance supercapacitors, *Journal of Science: Advanced Materials and Devices*, 2023, 100568.
2. Le Hong Quan, Ung Thi Dieu Thuy, Nguyen Van Chi, Nguyen Van Hoa, Chitosan derived N-doped carbon aerogel nanostructures for high-performance supercapacitors, *Advances in Natural Sciences: Nanoscience and Nanotechnology*, 2024, 045001.
3. Le Hong Quan, Ung Thi Dieu Thuy, Nguyen Van Chi, Nguyen Van Hoa, Chitosan-derived carbon and NiCo<sub>2</sub>O<sub>4</sub> aerogel composite for high-performance supercapacitors, *International Journal of Biological Macromolecules*, 2024, 136846
4. Le Hong Quan, Nguyen Van Hoa, Ung Thi Dieu Thuy, Nguyen Van Chi, The electrochemical performance of chitosan-derived carbon electrode material in aqueous electrolytes, *Proceedings of IWAMSN*, 22-25 September 2024, Da Nang, NMD-P08, 136-140.