# Supporting Information

## Role of pH in Tailoring Ni-Co Hydroxide Nanostructures for Energy Storage Applications

Rajkamal Arya[a*], Tanya Dagar[b], Arpit Thomas [c,d], M.N Singh[e], A. K. Sinha [a*]
Harpreet Singh Arora[c*]

[a*]Department of Physics, School of Advanced Engineering, UPES, Dehradun, Uttarakhand-248007, India

[b]Department of Chemistry, School of Advanced Engineering, UPES, Dehradun, Uttarakhand-248007, India

[c]Department of Mechanical Engineering, Shiv Nadar Institution of Eminence, deemed to be University, India (UP)- 201310

[d]Centre for Inter-Disciplinary Research and Innovation, University of Petroleum and Energy Studies, Bidholi Via-Prem Nagar, Dehradun-248007

[e]Accelerator Physics and Synchrotron Utilization Division, Raja Ramanna Centre for Advanced Technology, Indore 452013, India

***Table S1****: Comparing NCH with Known Supercapacitor Electrode Materials*

| **Materials** | **Synthetic route** | **Specific Capacitance (F $g^{-1}$)** | **Current Density (A $g^{-1}$)** | **Capacitance Retention at High Rate (%)** | **References** |
|---|---|---|---|---|---|
| $NiCo(OH)_4$ (NCH-2) | Co-precipitation/Hydrothermal | 621 | 0.5 | 64 % at 10 A $g^{-1}$ | This Work |
| $NiCo_2O_4$ Nanorods | Hydrothermal | 435 | 1.0 | 83.4 % at 1A $g^{-1}$ | [1] |
| Ni-Co LDHs | Electrodeposition | 690 | 1.0 | 60 % at 10 A $g^{-1}$ | [2] |
| $Cu(OH)_2$@NiCo-OH hierarchical | Electrodeposition + wet chemical | 770 | 5.0 | 95.8% over 6000 cycles | [3] |
| Ni-Fe -LDH | Co-precipitation | 1368 | 1.0 | 87.5% over 5000 cycles | [4] |
| Ni/Co hydroxide-(oxy)hydroxide | Co-precipitation | 566 | 1.0 | 84 % over 10000 cycles | [5] |

***Table S2:*** *Phase Composition and Structural Parameters of NCH Samples.*

| **Samples** | **α-phase Concentration (%)** | **β-phase Concentration (%)** | **α-phase Particle Size (nm)** | **β-phase Particle Size (nm)** | **β-phase micro-strain (%)** |
|---|---|---|---|---|---|
| NCH-1 | 37.5 | 62.5 | 12.6 | 45.4 | 0.145 |
| NCH-2 | 12.8 | 87.2 | 15.3 | 32.0 | 0.063 |
| NCH-3 | 26.0 | 74.0 | 15.8 | 32.8 | 0.069 |
| NCH-4 | 94.9 | 5.1 | 10.2 | 31.9 | - |

***Table S3:*** *BET and BJH Surface Area, Pore Volume of NCH Samples.*

| **Samples** | **Pore Volume ($cm^3/g$)** | **Specific Surface Area (BET) ($m^2/g$)** | **Specific Surface Area (BJH) ($m^2/g$)** |
|---|---|---|---|
| NCH-1 | 0.058 | 22.11 | 28.03 |
| NCH-2 | 0.055 | 12.83 | 13.86 |
| NCH-3 | 0.186 | 42.55 | 48.50 |
| NCH-4 | 0.297 | 52.32 | 59.54 |

***Table S4:*** *Kinetic parameters and charge storage contributions (SCP and DIP) of NCH samples obtained from CV analysis.*

| **Sample** | **Process** | **b-value** | **$b_2$ (Capacitive)** | **$b_1$ (Diffusion)** | **SCP (%)** | **DIP (%)** |
|---|---|---|---|---|---|---|
| NCH-1 | Anodic | 0.50 | 0.99 ± 0.01 | - | - | 100 |
| NCH-1 | Cathodic | - | - | - | - | - |
| NCH-2 | Anodic | 0.61 | 0.82 ± 0.06 | 3.18 ± 0.16 | 20 | 80 |
| NCH-2 | Cathodic | 0.56 | 0.86 ± 0.06 | 2.88 ± 0.18 | 23 | 77 |
| NCH-3 | Anodic | 0.60 | 0.70 ± 0.05 | 3.92 ± 0.18 | 15 | 85 |
| NCH-3 | Cathodic | 0.57 | 0.74 ± 0.05 | 3.48 ± 0.19 | 17 | 83 |

| | | | | | | |
|---|---|---|---|---|---|---|
| NCH-4 | Anodic | 0.64 | 0.62 ± 0.05 | 4.70 ± 0.20 | 12 | 88 |
| NCH-4 | Cathodic | 0.64 | 0.58 ± 0.06 | 3.65 ± 0.28 | 14 | 86 |

***Table S5:*** *Specific capacitance of NCH samples at various current densities.*

| **Current Density (A/g)** | **NCH-1 (F/g)** | **NCH-2 (F/g)** | **NCH-3 (F/g)** | **NCH-4 (F/g)** |
|---|---|---|---|---|
| 0.5 | - | 621 | 480 | 343 |
| 1 | 388 | 552 | - | 127 |
| 2 | 314 | 506 | 440 | 110 |
| 3 | - | - | 366 | 105 |
| 5 | 161 | 462 | 313 | 88 |
| 6 | - | 405 | - | - |
| 10 | 110 | 400 | 250 | 75 |
| 15 | 98 | - | 225 | 38 |

***Table S6:*** *Electrochemical impedance spectroscopy (EIS) fitting parameters of NCH samples obtained using the equivalent circuit model, along with chi-square ($\chi^2$) values indicating fitting quality.*

| **Samples** | **$R_s$ (Ω)** | **$R_{ct}$ (Ω)** | **CPE-T** | **CPE-P** | **$Z_{Wo\text{-}R}$ (Ω)** | **$Z_{Wo\text{-}T}$** | **$Z_{Wo\text{-}P}$** | **Chi-square error ($\chi^2$)** |
|---|---|---|---|---|---|---|---|---|
| NCH-1 | 0.50 | 4.73 | 0.00258 | 0.41 | 32.3 | 48.8 | 0.66 | $6.7\times 10^{-4}$ |
| NCH-2 | 0.21 | 0.38 | 0.07 | 0.72 | 57 | 44 | 0.51 | $4.2\times 10^{-4}$ |
| NCH-3 | 0.45 | 0.52 | 0.50 | 0.97 | 70 | 60 | 0.78 | $6.9\times 10^{-3}$ |
| NCH-4 | 0.42 | 4.30 | 0.36 | 0.93 | 50 | 61 | 0.80 | $8.2\times 10^{-4}$ |

***Table S7:*** *Figure 1 Specific capacitance, energy density, and power density of the NCH-2 symmetric coin cell device at different current densities.*

| **Current Density (A $g^{-1}$)** | **Specific Capacitance (F $g^{-1}$** | **Energy Density (Wh $kg^{-1}$)** | **Power Density (W $kg^{-1}$** |
|---|---|---|---|
| 0.25 | 22.0 | 6.86 | 178 |
| 0.50 | 20.6 | 6.43 | 618 |
| 0.75 | 17.5 | 5.47 | 547 |
| 1.00 | 10.0 | 3.13 | 714 |

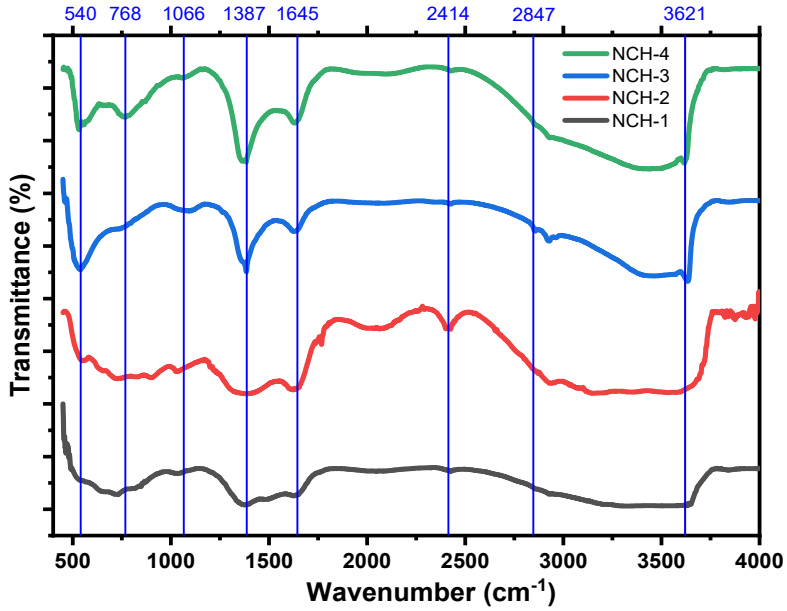



***Figure S1****: FTIR Spectrum of NCH samples*

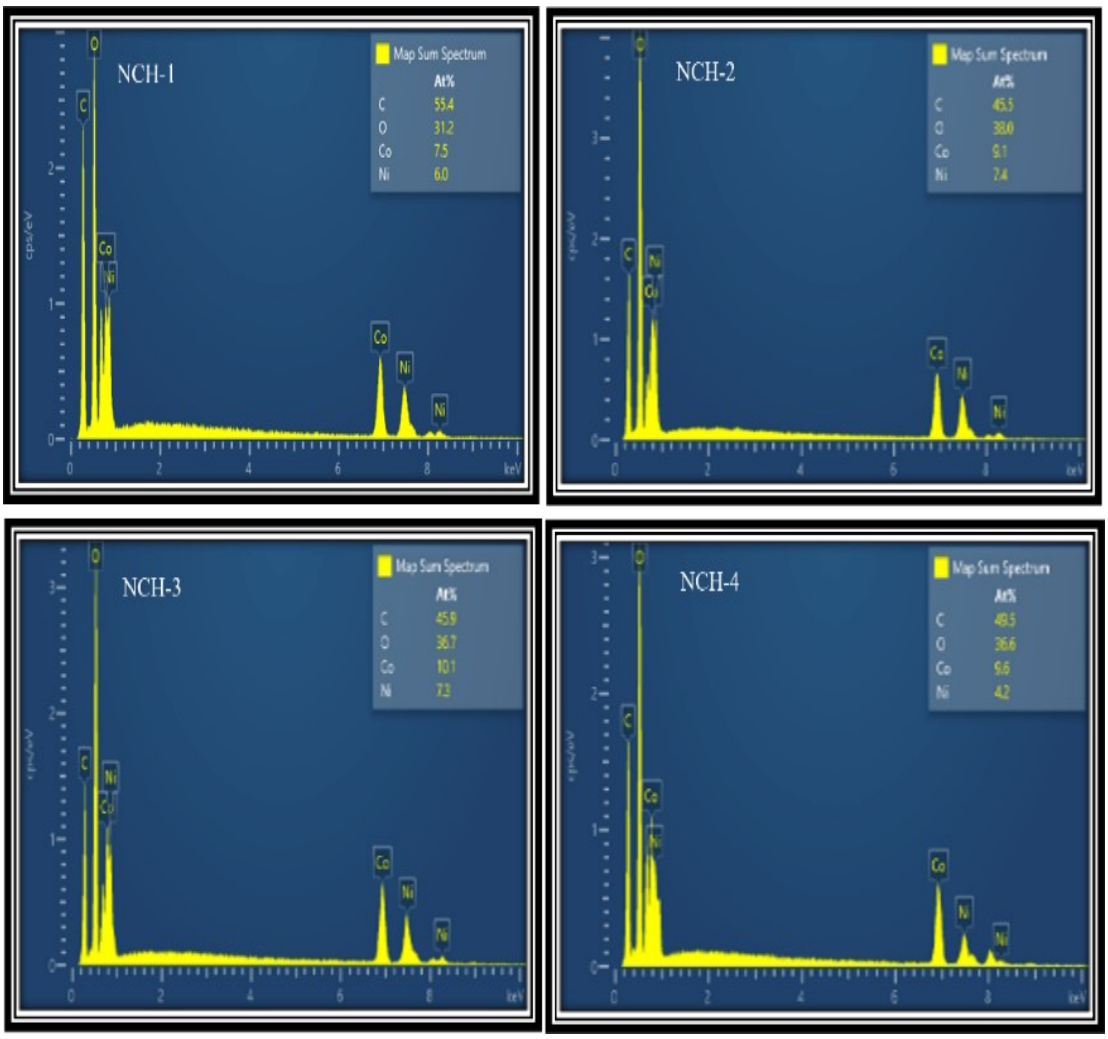



***Figure S2:*** *EDX elemental mapping images of NCH samples.*

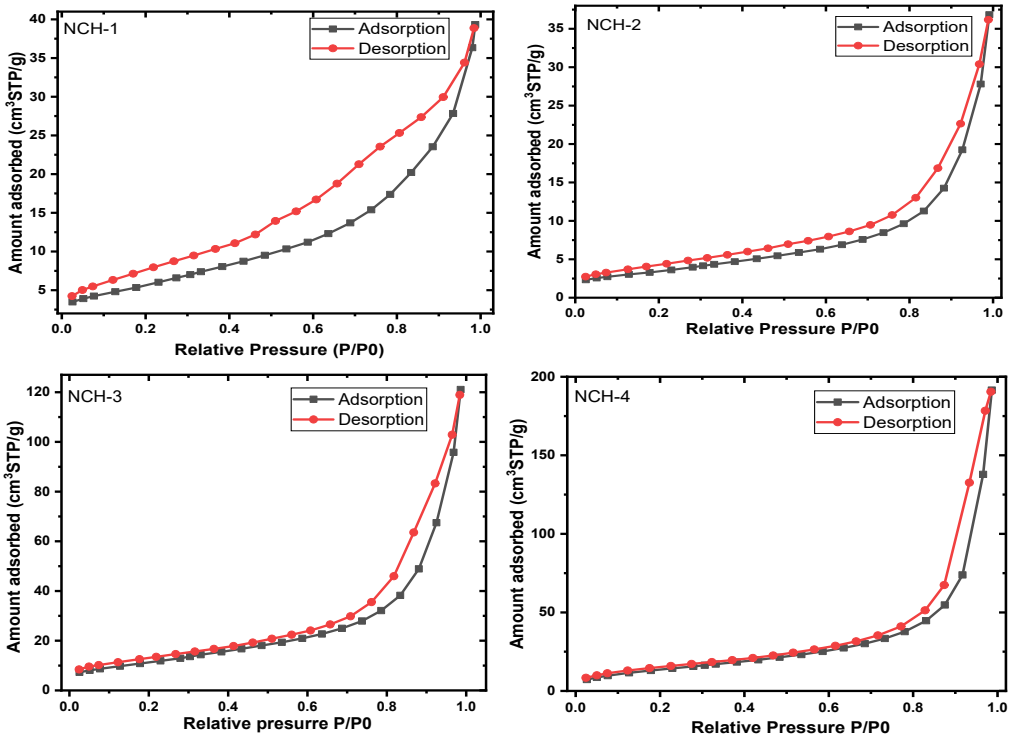



***Figure S3**: Adsorption-desorption isotherms plot of NCH samples*

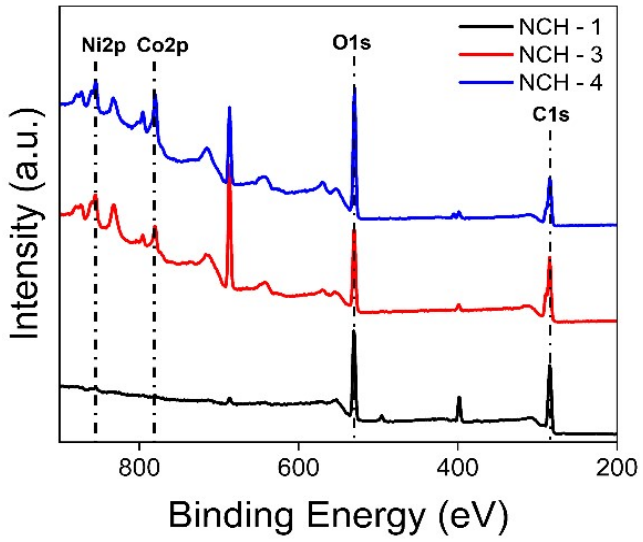



***Figure S4:** XPS Survey scan of NCH-1,3, 5 samples*

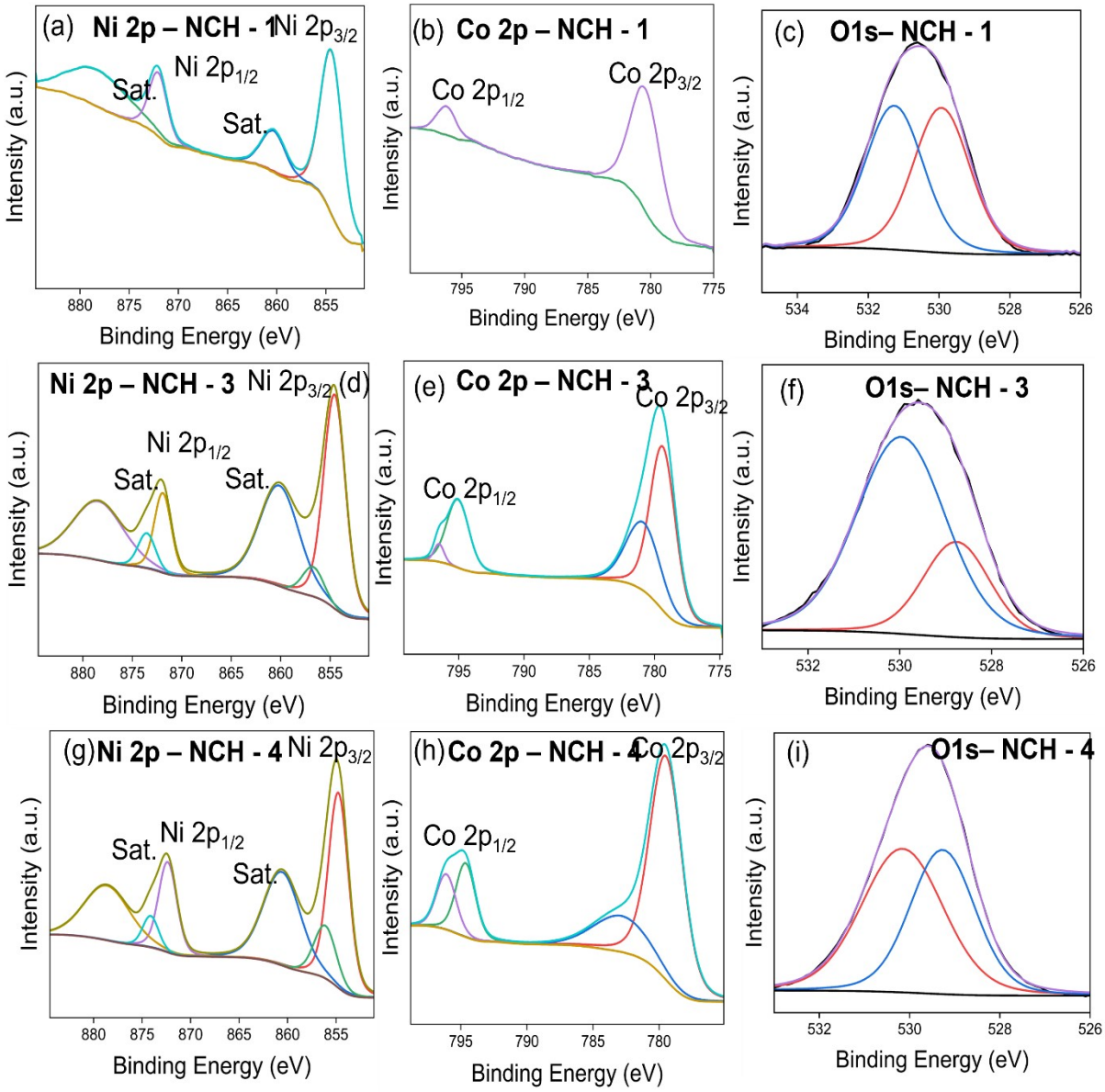



***Figure S5:*** *XPS spectrum of (a-c) NCH-1, (d-f) NCH-3, (g-i) NCH-4 samples respectively*

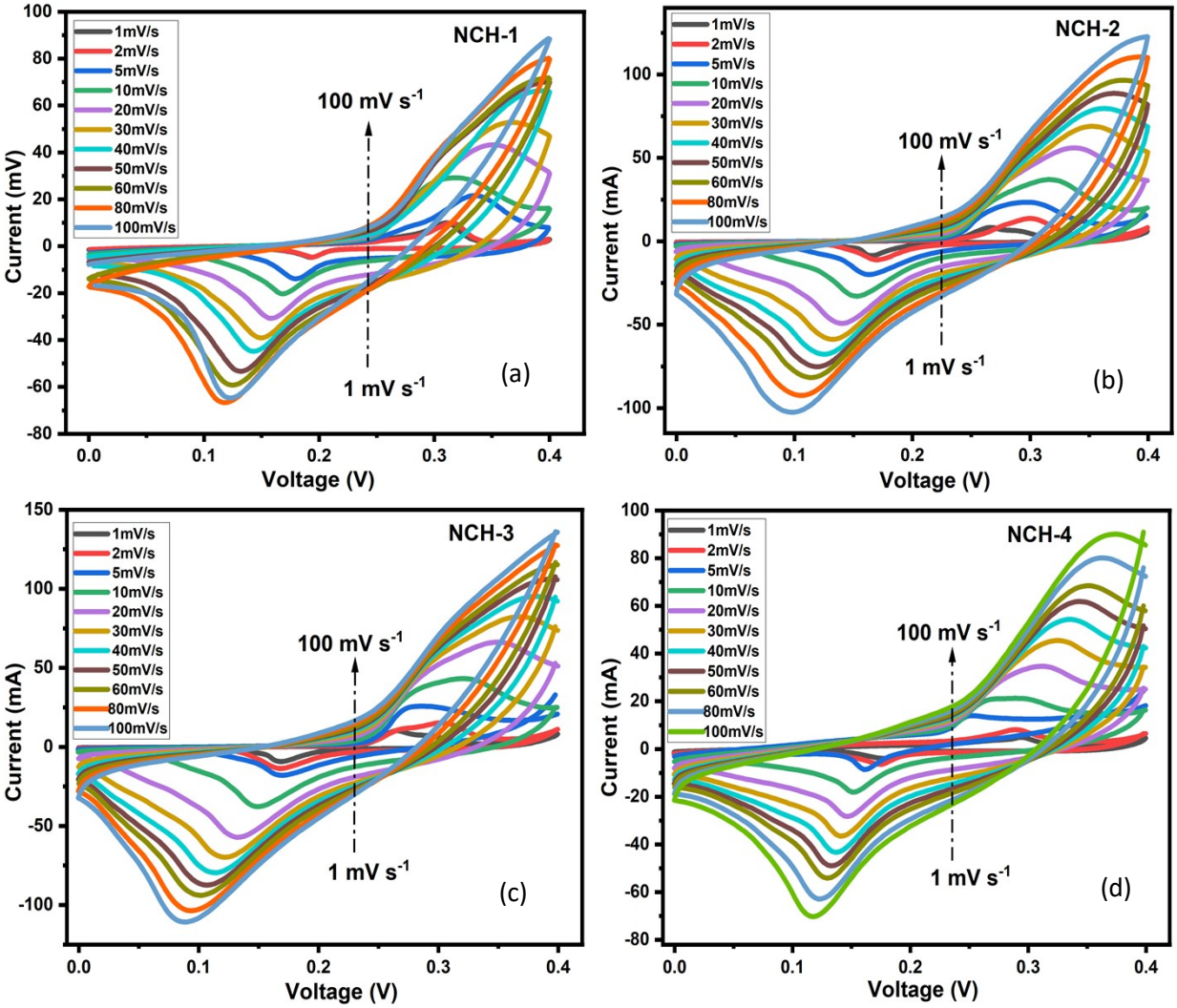



***Figure S6:*** *(a-d) Cyclic voltammetry plot of NCH samples from 1 to 100 mV/s.*

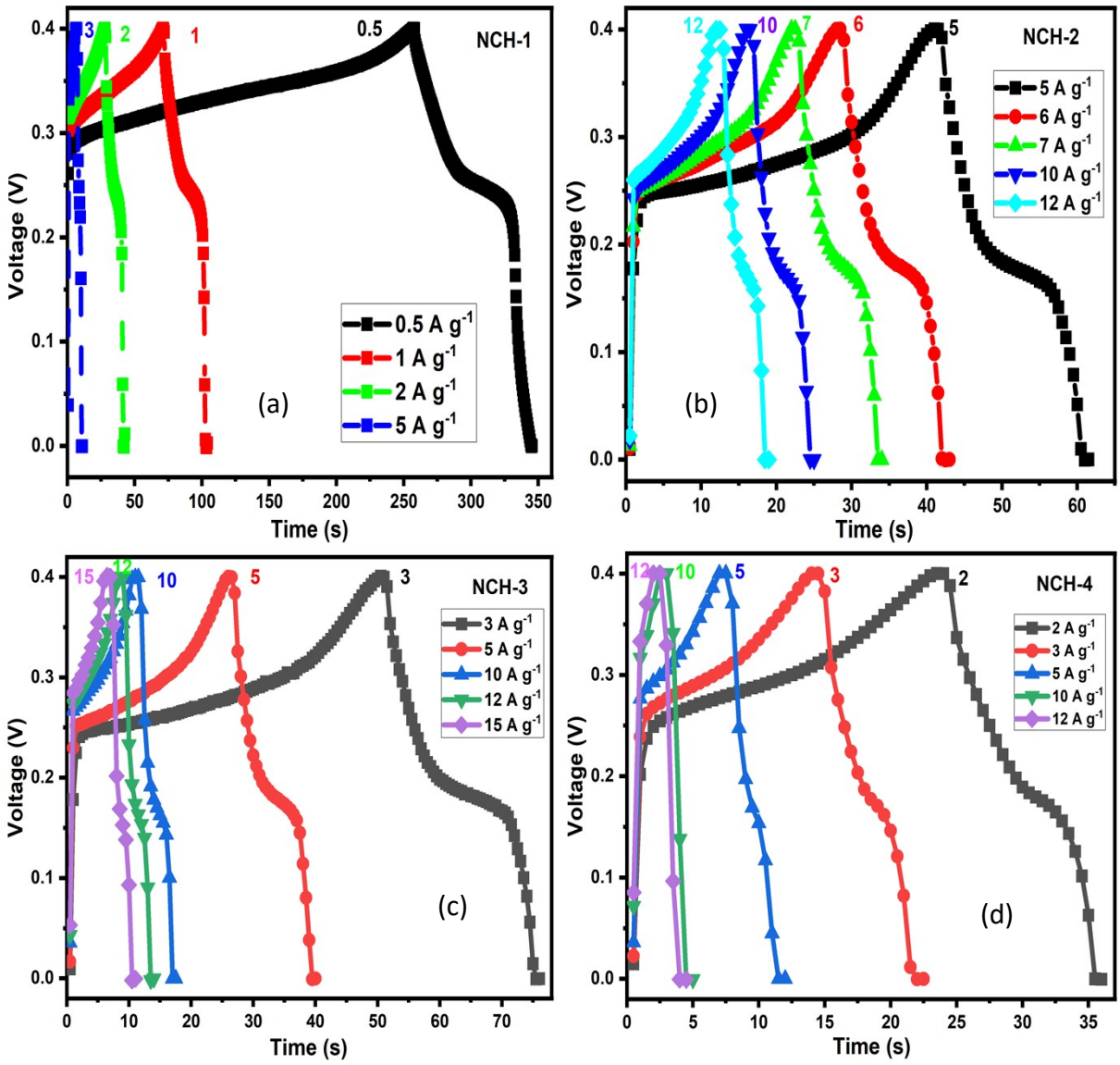
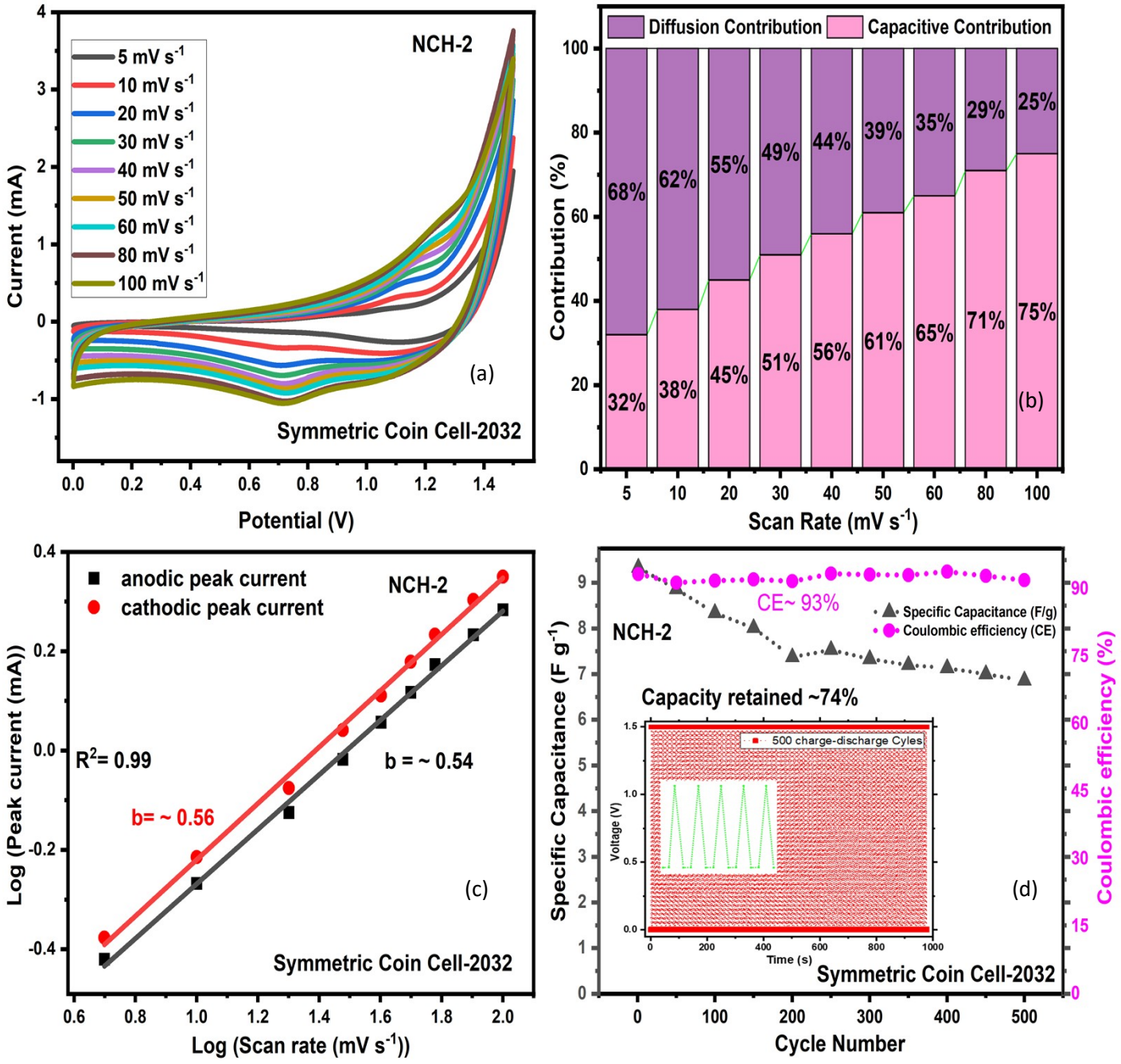



***Figure S7:*** *GCD plot of NCH samples at various current densities*




***Figures S8:*** *Electrochemical performance of symmetric NCH-2 Coin Cell-2032 device (a) CV from various scan rates, (b) Capacitive-Diffusive controlled process in supercapacitor, (c) Log-log plot between Peak current and Scan rate, (d) Cyclic stability, and Coulombic efficiency. (Inset: 500 Charge-discharge cycles)*

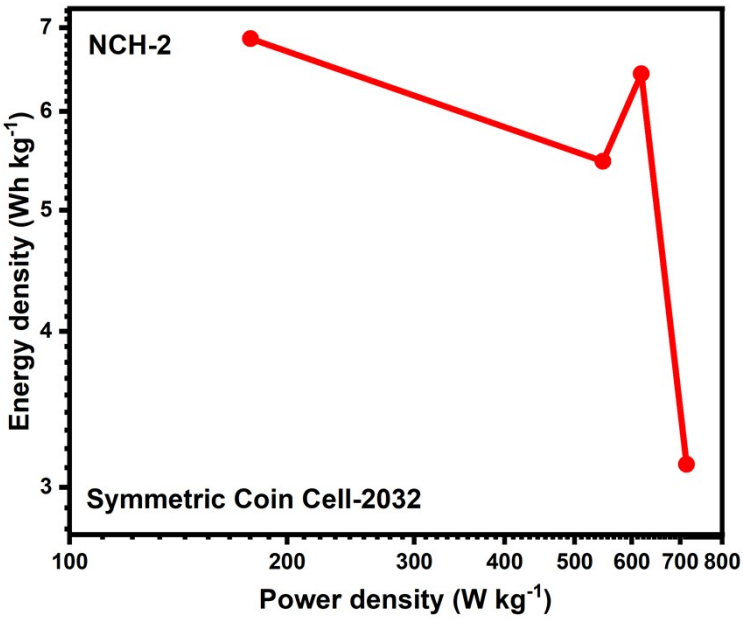



***Figure S9:*** *Ragone plot of the NCH-2 symmetric CR-2032 device.*

**References:**


1 S. Savithri, P. Remya, S. Vanitharaj, S. Selvakumar, P. Krishnan, S. Sudharthini, V. P. Venkatesh, R. Balaji and R. Nithya, *Chem. Phys. Lett.*, 2025, **869**, 142028.

2 C. Xu, J. Li, K. Yang, X. Zhao and M. Liu, *J. Mater. Chem. A Mater.*, 2014, **2**, 2162–2168.

3 X. Su, H. Shi, Y. Xu, D. Zhang, Z. Guo, J. Yu, J. Sui, Q. Zhang, L. Yu and L. Dong, *Journal of Electroanalytical Chemistry*, 2026, **1000**, 119593.

4 B. R. Wiston, P. Prabhakaran and M. Ashok, *J. Energy Storage*, 2022, **50**, 104226.

5 A. Roy, H. R. Inta, S. Ghosh, H. V. S. R. M. Koppisetti, A. Mondal, B. R. Verma, S. Bag and V. Mahalingam, *J. Mater. Chem. A Mater.*, 2024, **12**, 4086–4098.