Impact of Neodymium Doping on Nitrogen Enriched Co/CoS for High-Performance Dye-Sensitized Solar Cells and Supercapacitors

Imani Sospeter[a,c], Manik Clinton Franklin[a], Hemalatha Kuzhandaivel[b], Karthick Sivalingam Nallathambi[a]*

[a] *Electrochemical Materials & Devices Lab, Department of Chemistry, Bharathiar University, Coimbatore-641046, Tamil Nadu, India.*

[b] *Materials Research & Product Laboratory, Department of Chemistry, Coimbatore Institute of Technology, Coimbatore-641014, Tamil Nadu, India.*

[c]*Department of Applied Science and Social Studies, Arusha Technical College, P.O. Box 296, Arusha,* Tanzania

*Corresponding author e-mail: snkarthick@buc.edu.in

Supporting information

Table S1. Elemental composition of the core-shell NCS4 and CS samples measured by XPS analysis.

Sample	Co (at. %)		S (at. %)	N (at. %)	Nd (at. %)
	$Co^{(0)}$	Co^{2+}			
	7.45	14.28			
NCS4	**21.73**		**54.12**	**21.55**	**3.75**
	6.3	16.9			
CS	**23.32**		**53.79**	**21.87**	**-**

Table S2: Comparison of the performance of the already reported counter electrodes and the Nd-doped-N-Co/CoS counter electrode used in DSSC.

S/No	Materials	Methods	Electrolyte	PCE (%)	Ref.
1	$CoNi_2S_4$	Spin coating	I^-/I_3^-	3.45	[1]
2	Ni-$MoSe_2$	Doctor blade	I^-/I_3^-	6.78	[2]
3	CoS_2	Doctor blade	I^-/I_3^-	6.13	[3]
4	$Co_{0.42}Ni_{0.58}Se$	Doctor blade	I^-/I_3^-	6.15	[4]
5	CoS	Doctor blade	I^-/I_3^-	4.74	[5]

6	WS_2	Doctor blade	I^-/I_3^-	4.4	[6]
7	$NiAl_2S_4$	Doctor blade	I^-/I_3^-	5.02	[7]
8	MoS_2	Grown-FTO	I^-/I_3^-	6.22	[8]
9	NCS4	Doctor blade	I^-/I_3^-	6.90	**This work**

Table S3: Comparison of the already reported electrode material with the NCS4 electrode material used in SSC.

S/N	**Electrode materials**	**Electrolyte**	**Specific capacitance (F/g)**	**Specific energy (Wh/kg)**	**Specific power (kW/kg)**	**Ref.**
1	NiS	PVA-KOH	11.15 at 1 A/g	0.99	0.132	[9]
2	$CuFeS_2$	1M LiOH	34.18 at 1 A/g	4.74	0.166	[10]
3	Yb_2S_3	1M Na_2SO_4	23 at 0.5 A/g	8.25	0.56	[11]
4	$MoSe_2$ /MWCNT	1 M KOH	27.2 at 0.4 A/g	7.41	1.4	[12]
5	Dy_2S_3	PVA-Na_2SO_4	26 at 2.1 A/g	17	0.52	[13]
6	1T-MoS_2	0.5 M k_2SO_4	37.2 at 0.5 A/g	4.19	0.225	[14]
7	CuS/MnS@NF	2 M KOH	37.4 at 0.5A/g	2.54	0.17	[15]
8	NCS4	3 M KOH	66.35 at 1 A/g	23.6	1.6	**This work**

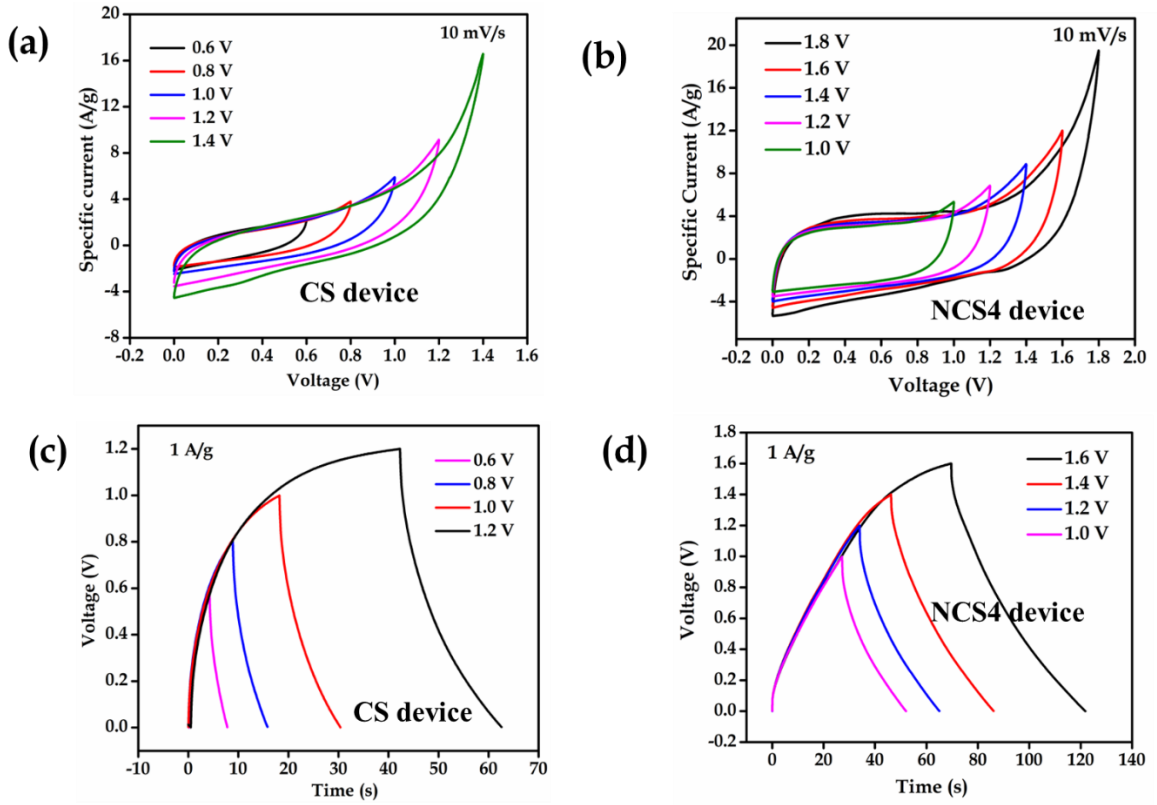

Fig. S1: Comparison of CS and NCS4 symmetric supercapacitor devices using different potential windows, CV plots, and GCD curves.

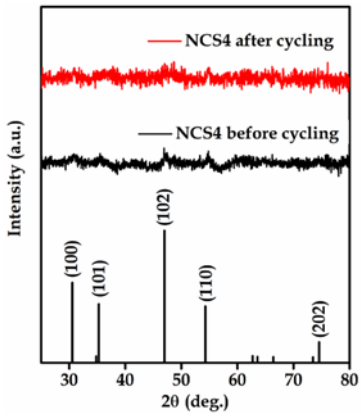

Fig. S2: XRD pattern of the NCS4 compounds before and after 2000 charge-discharge cycles.

References

(1) Sankar, G., Anbarasu, P., Mahendran, R., Rajendran, K., Sivakumar, T. Pt-free and efficient counter electrode with nanostructured $CoNi_2S_4$/rGO for dye-sensitized solar cells. *Inorganic Chemistry Communications*, **2021,** *126*, 108475.

(2) Althomali, R. H., Al-Hawary, S. I. S., Gehlot, A., Qasim, M. T., Abdullaeva, B., Sapaev, I. B., Alsalamy, A. A novel Pt-free counter electrode based on $MoSe_2$ for cost effective dye-

sensitized solar cells (DSSCs): Effect of Ni doping. *Journal of Physics and Chemistry of Solids*, **2023**, *182*, 111597.

(3) Tsai, J. C., Hon, M. H., Leu, I. C. Fabrication of Mesoporous CoS_2 Nanotube Arrays as the Counter Electrodes of Dye-Sensitized Solar Cells. *Chemistry–An Asian Journal*, **2015**, *10*(9), 1932-1939.

(4) Jiang, Q., Pan, K., Lee, C. S., Hu, G., Zhou, Y. Cobalt-nickel based ternary selenides as high-efficiency counter electrode materials for dye-sensitized solar cells. *Electrochimica Acta*, **2017,** *235*, 672-679.

(5) Wang, G., Zhang, J., Kuang, S., Liu, S., Zhuo, S. The production of cobalt sulfide/graphene composite for use as a low-cost counter-electrode material in dye-sensitized solar cells. *Journal of Power Sources*, **2014**, *269*, 473-478.

(6) Wang, Y., Li, S., Bai, Y., Chen, Z., Jiang, Q., Li, T., Zhang, W. Dye-sensitized solar cells based on low-cost carbon-coated tungsten disulphide counter electrodes. *Electrochimica Acta*, **2013**, *114*, 30-34

(7) Tamilarasan, S., Sundararaj, S. B., Bhojanaa, K. B., Pandikumar, A., Basha, S. S., Thangavelu, S. Unraveling the enhanced electrochemical performances of nickel aluminum sulfide lattices for overall water splitting and as counter electrode materials for dye-sensitized solar cells. *The Journal of Physical Chemistry C*, **2023**, *127*(25), 11921-11931.

(8) Hu, Z., Xia, K., Zhang, J., Hu, Z., Zhu, Y. Highly transparent ultrathin metal sulfide films as efficient counter electrodes for bifacial dye-sensitized solar cells. *Electrochimica Acta*, **2015**, *170*, 39-47.

(9) Parveen, N., Ansari, S. A., Ansari, S. G., Fouad, H., Abd El-Salam, N. M., Cho, M. H. Solid-state symmetrical supercapacitor based on hierarchical flower-like nickel sulfide with shape-controlled morphological evolution. *Electrochimica Acta*, **2018,** *268*, 82-93.

(10) Sahoo, S., Pazhamalai, P., Mariappan, V. K., Veerasubramani, G. K., Kim, N. J., Kim, S. J. Hydrothermally synthesized chalcopyrite platelets as an electrode material for symmetric supercapacitors. *Inorganic Chemistry Frontiers*, **2020**, *7*(7), 1492-1502.

(11) Ubale, S. B., Kale, S. B., Mane, V. J., Bagwade, P. P., Lokhande, C. D. SILAR synthesized nanostructured ytterbium sulfide thin film electrodes for symmetric supercapacitors. *Journal of Solid-state electrochemistry*, **2021,** *25*(6), 1753-1764.

(12) Karade, S. S., Sankapal, B. R. Two dimensional cryptomelane like growth of $MoSe_2$ over MWCNTs: symmetric all-solid-state supercapacitor. *Journal of Electroanalytical Chemistry*, **2017**, *802*, 131-138.

(13) Bagwade, P. P., Nikam, R. P., Bhosale, R. P., Khot, S. D., Lokhande, C. D. Performance of solid-state symmetric supercapacitors based on Dy_2S_3 electrodes. *Applied Surface Science Advances*, **2023,** *18*, 100529.

(14) Ali, B. A., Omar, A. M., Khalil, A. S., Allam, N. K. Untapped potential of polymorph MoS_2: tuned cationic intercalation for high-performance symmetric supercapacitors. *ACS applied materials & interfaces*, **2019**, *11*(37), 33955-33965.

(15) Zhai, M., Cheng, Y., Jin, Y., Hu, J. Solvothermal synthesis of flower-like structure Cu-Mn bimetallic sulfide on Ni-foam for high-performance symmetric supercapacitors. *International Journal of Hydrogen Energy*, **2019**, *44*(26), 13456-13465.