Unveiling the potential of gadolinium-doped nitrogen-enriched Co/CoS as an efficient electrode for dye-sensitized solar cells and supercapacitors applications

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Supporting information

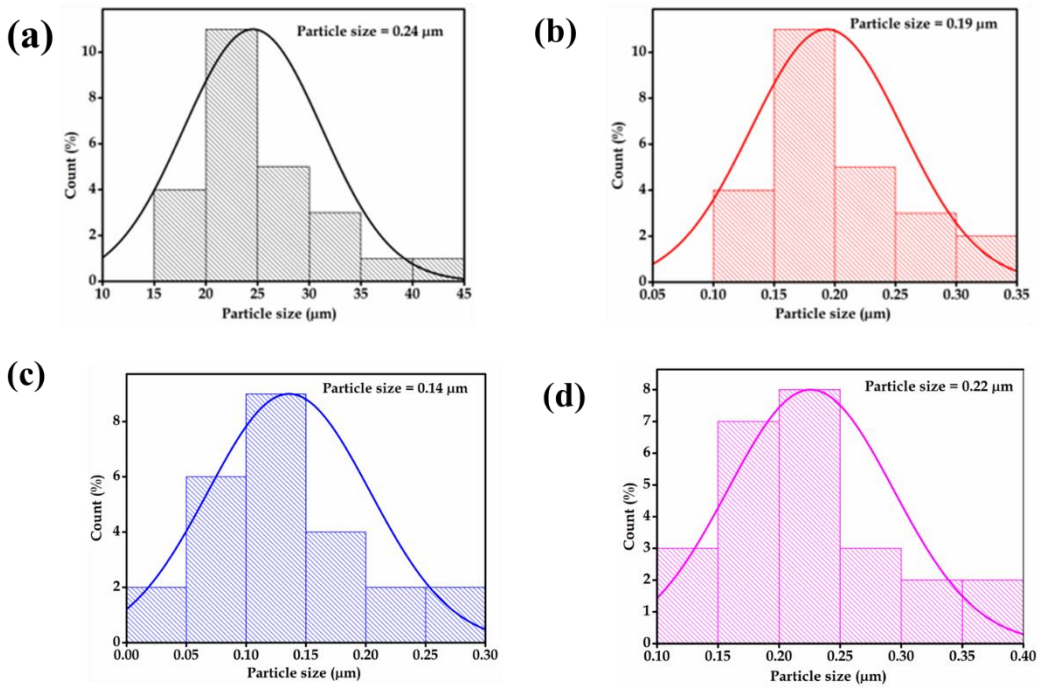

Fig. S1. Histogram of the average particle size distribution of (a) CS (b) GCS2 (c) GCS4 (d) GCS6.

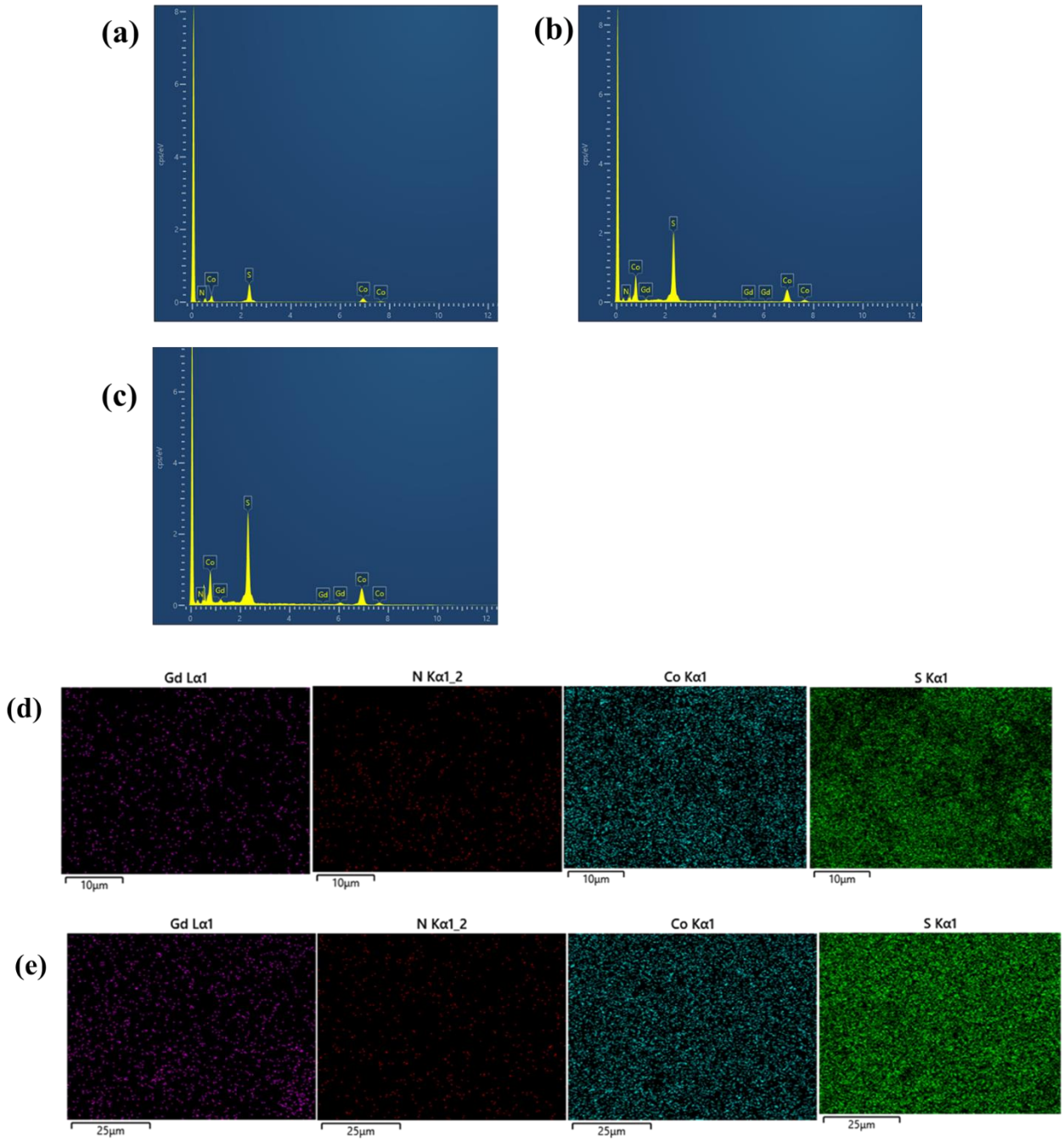

Fig. S2 SEM-EDS spectra and mapping images of **(a)** CS **(b, d)** GCS2 **(c, e)** GCS6

Table S1: Comparison of the performance of the already reported DSSCs using metal chalcogenide counter electrodes and the fabricated DSSC using 4% Gd-doped -N-Co/CoS counter electrode.

S/No	Materials	Methods	Electrolyte	PCE (%)	Ref.
1	$CuInS_2$	Doctor blade	I^-/I_3^-	5.79	[1]
2	$NiAl_2S_4$	Doctor blade	I^-/I_3^-	5.02	[2]
3	MoS_2	Grown on the FTO	I^-/I_3^-	6.22	[3]
4	FeS_2	Doctor blade	I^-/I_3^-	5.93	[4]
5	$Co_{0.42}Ni_{0.58}Se$	Doctor blade	I^-/I_3^-	6.15	[5]
6	MoS_2	Electrodeposition	I^-/I_3^-	6.89	[6]
7	Ni-$MoSe_2$	Doctor blade	I^-/I_3^-	6.78	[7]
8	CoS	Electrodeposition	I^-/I_3^-	7.16	[8]
9	GCS4	Doctor blade	I^-/I_3^-	7.3	**This work**

Table S2: Benchmark of the already reported SSC device compared to the GCS4 SSC device.

S/N	Electrode materials	Electrolyte	Specific capacitance (F/g)	Specific energy (Wh/kg)	Specific power (kW/kg)	Ref.
1	Mn-MoS_2	0.5 M Na_2SO_4	88 at 1 A/g	48.9	5.0	[9]
2	Gd-MoS_2	0.5 M Na_2SO_4	88 at 0.5 A/g	30	1.7	[10]
3	MnS	PVA-KOH	68 at 1A/g	18.90	3.3	[11]
4	MSe/Mo_3Se_4	1.0 M KOH	89 at 1 A/g	29.3	2.0	[12]
5	3D graphene/MoS_2	1.0 M KOH	58 at 1 A/g	24.59	1.00	[13]
6	Co-MoS_2	1 M KOH	86 at 1.0 A/g	4.3	0.6	[14]
7	MoS_2-GO	PEI-GO	42.9 at 1 A/g	19.3	4.5	[15]
8	GCS4	3 M KOH	92.10 at 1 A/g	32.74	1.69	**This work**

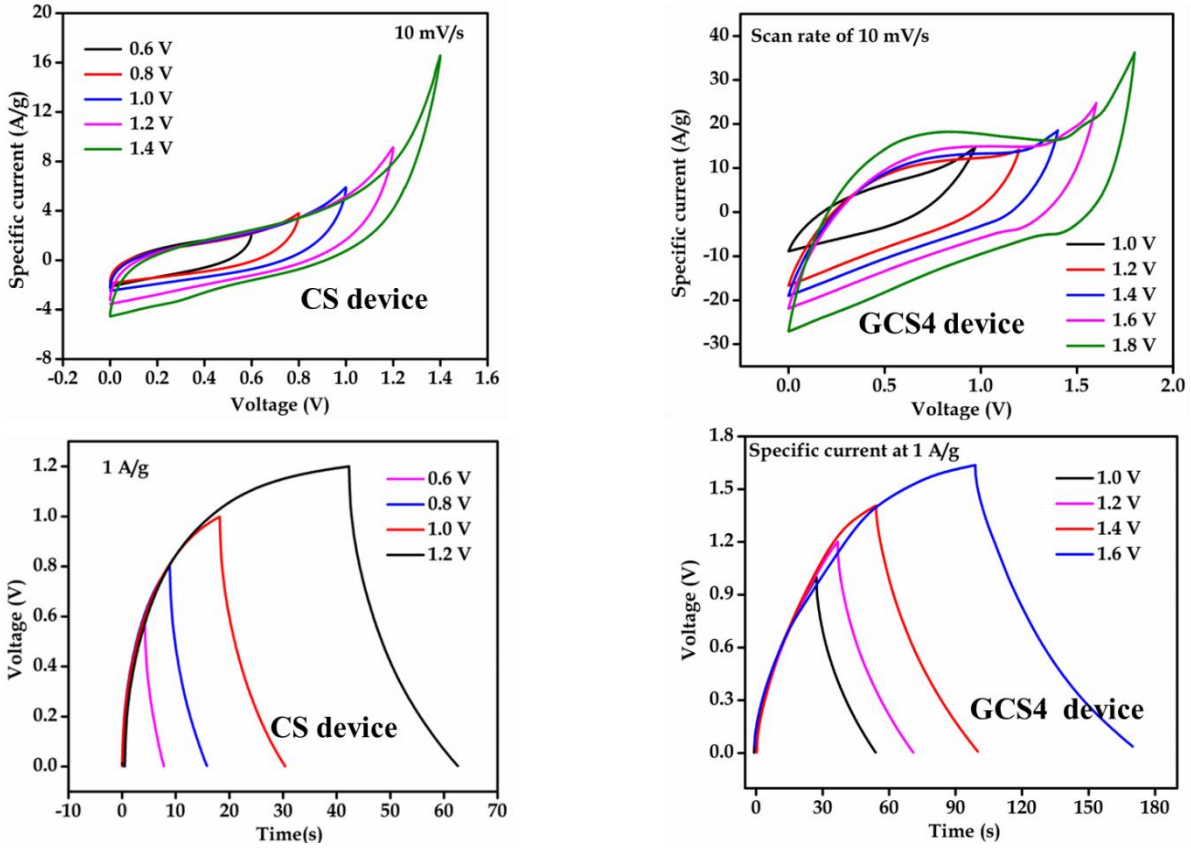

Fig. S3: Comparison of CS and GCS4 symmetric supercapacitor devices using different potential windows for CV plots and GCD curves.

References

1. Das, S., Banerjee, A., Pal, P., Rudra, S., Nandi, U., Ghosh, A. Hydrothermally synthesized gadolinium doped molybdenum disulfide for electrochemical supercapacitor applications. *Journal of Energy Storage*, 2024, *99*, 113268.
2. Wang, L., He, J., Zhou, M., Zhao, S., Wang, Q., Ding, B. Copper indium disulfide nanocrystals supported on carbonized chicken eggshell membranes as efficient counter electrodes for dye-sensitized solar cells. *Journal of Power Sources*, 2016, *315*, 79-85.
3. 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.

4. 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.
5. Song, C., Wang, S., Dong, W., Fang, X., Shao, J., Zhu, J., Pan, X. Hydrothermal synthesis of iron pyrite (FeS2) as efficient counter electrodes for dye-sensitized solar cells. *Solar Energy, 2016*, *133*, 429-436.
6. 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. *Electrochim. Acta* , 2017, 235, 672–679.
7. Li, S., Min, H., Xu, F., Tong, L., Chen, J., Zhu, C., Sun, L.. All electrochemical fabrication of MoS_2/graphene counter electrodes for efficient dye-sensitized solar cells. *RSC Advances*, 2016, *6*(41), 34546-34552.
8. 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.
9. Xiao, Y., Han, G., Chang, Y., Zhang, Y., Lin, J. Y. Cobalt sulfide counter electrodes enhanced by a hydro-thermal treatment for use in platinum-free dye-sensitized solar cells. *Materials Research Bulletin*, 2015, 68, 9-15.
10. Singha, S. S., Rudra, S., Mondal, S., Pradhan, M., Nayak, A. K., Satpati, B., Singha, A. Mn incorporated MoS_2 nanoflowers: A high-performance electrode material for symmetric supercapacitor. *Electrochimica Acta*, 2020, *338*, 135815.
11. Pujari, R. B., Lokhande, A. C., Yadav, A. A., Kim, J. H. Lokhande, C. D. Synthesis of MnS microfibers for high performance flexible supercapacitors. *Materials & Design*, 2016, *108*, 510-517.
12. Krishna, B. V., Ankinapalli, O. R., Reddy, A. R., Yu, J. S. Facile one-step hydrothermal route to MSe/Mo_3Se_4 (M: Zn, Mn, and Ni)-based electrode materials for ultralong-life hybrid supercapacitors. *Journal of Materials Science & Technology*, 2023, *156*, 230-240.
13. Singh, K., Kumar, S., Agarwal, K., Soni, K., Ramana Gedela, V., Ghosh, K. Three-dimensional graphene with MoS_2 nanohybrid as potential energy storage/transfer device. *Scientific reports*, 2017, *7*(1), 9458.

14. Rohith, R., Manuraj, M., Jafri, R. I., & Rakhi, R. B. Co-MoS_2 nanoflower coated carbon fabric as a flexible electrode for supercapacitor. *Materials Today: Proceedings*, 2022, *50*, 1-6.
15. Liu, M. C., Xu, Y., Hu, Y. X., Yang, Q. Q., Kong, L. B., Liu, W. W., Chueh, Y. L. Electrostatically charged MoS_2/graphene oxide hybrid composites for excellent electrochemical energy storage devices. *ACS applied materials & interfaces*, 2018, 10(41), 35571-35579.