Supplementary Information

Two Birds with One Stone: Cobalt/Silicon Species Encapsulated in MOF-derived Nitrogen-doped Carbon as an Integrated Electrode for Next-Generation Symmetric Pseudocapacitor with Energy Density over 100 Wh/kg

Abdul Mateen[a,1], Muhammad Sufyan Javed[b,,1], Xiaofeng Zhang[b,1], Iftikhar Hussain[c], Tayyaba Najam[d], Awais Ahmad[e], Asma A. Alothman[f], Mohamed Ouladsmane[f], Sayed M. Eldin[g], Weihua Han[b,*], Kui-Qing Peng[a,*]*

[a]Department of Physics and Beijing Key Laboratory of Energy Conversion and Storage Materials, Beijing Normal University, Beijing, 100084, China

Email: kq_peng@bnu.edu.cn

[b]School of Physical Science and Technology, Lanzhou University, Lanzhou 730000, China

Email: safisabri@gmail.com, hanwh@lzu.edu.cn

[c]Department of Mechanical Engineering, City University of Hong Kong, 83 Tat Chee Avenue, Kowloon, Hong Kong

[d]College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen 518060, China

[e]Departamento de Quimica Organica, Universidad de Cordoba, Edificio Marie Curie (C-3), 8 Ctra Nnal IV-A, Km 396, E14014 Cordoba, Spain

[f]Department of Chemistry, College of Science, King Saud University, Riyadh 11451, Saudi Arabia

[g]Faculty of Engineering and Technology, Future University in Egypt, New Cairo 11835, Egypt

[1]Equally contributed

Supporting Figures:

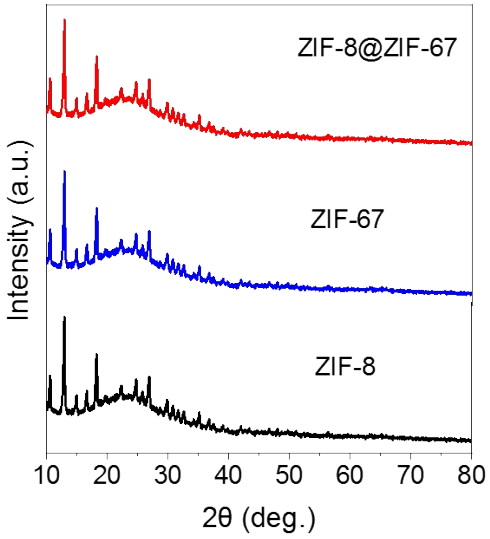

Figure S1: XRD patterns of core-shell ZIF-67@ZIF-8, ZIF-67 and ZIF-8

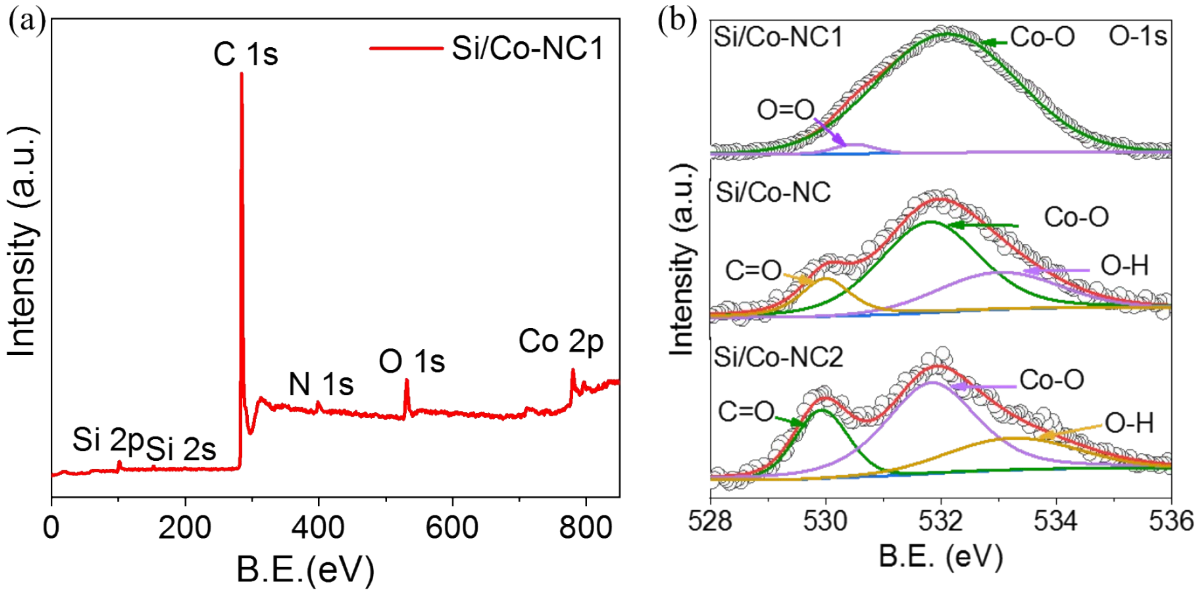

Figure S2: (a) Full XPS survey spectrum of Si/Co-NC1; (b) Deconvoluted XPS spectrums of O1s

Table S1: Average pore size of all the samples Si/CO-NC, Si/CO-NC1, Si/CO-NC2, Si-NC, and Co-NC.

Sample	Average pore size (nm)
Si/CO-NC	13.8
Si/CO-NC1	10.1
Si/CO-NC2	13.4
Si-NC	17. 2
Co-NC	26.5

Table S2: A comparison of charge transfer resistance (Rct (Ω)) and equivalent series resistance (Rs (Ω)) values of all the electrodes Si/CO-NC, Si/CO-NC1, Si/CO-NC2, Si-NC, and Co-NC.

Electrode	R_S (Ω)	R_{CT} (Ω)
Si/CO-NC	1.80	4.40
Si/CO-NC1	1.40	3.50
Si/CO-NC2	1.64	3.66
Si-NC	1.45	4.98
Co-NC	1.35	4.55

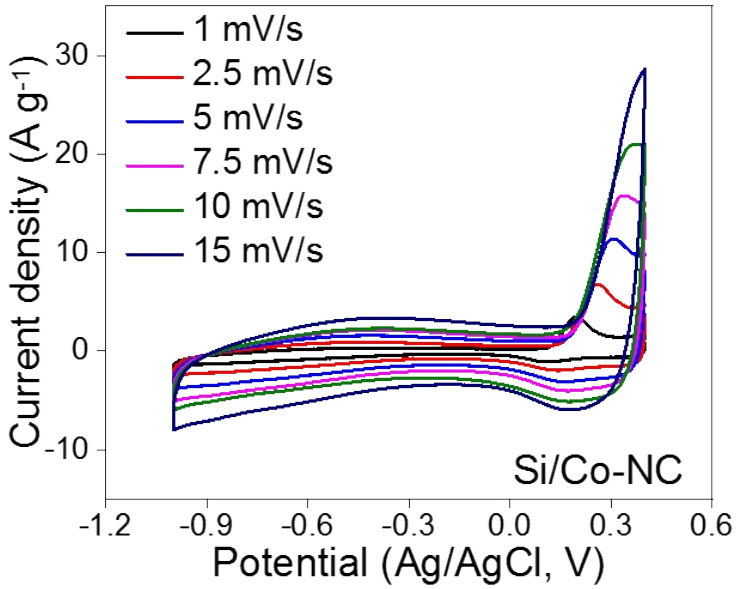

Figure S3: CV profiles of Si/Co-NC integrated electrode at various scan rates from 1 to 15 mV/s.

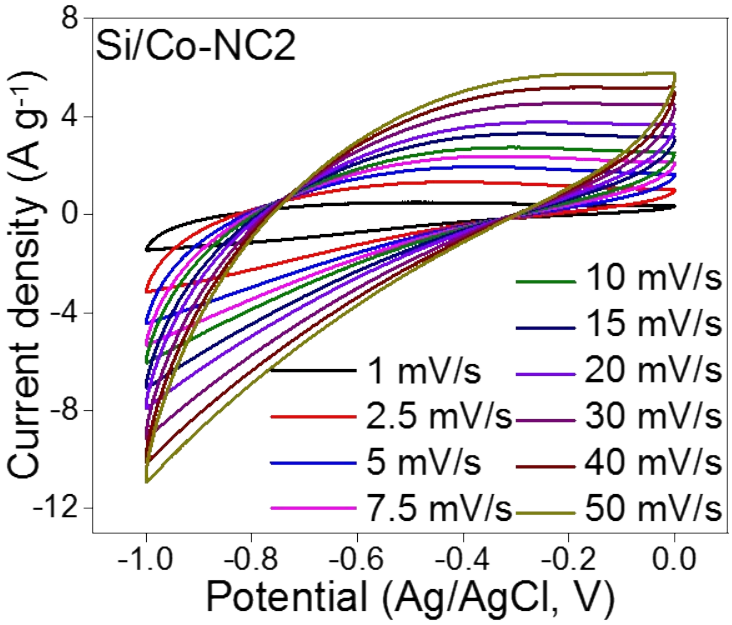

Figure S4: CV profiles of Si/Co-NC2 integrated electrode at various scan rates from 1 to 50 mV/s.

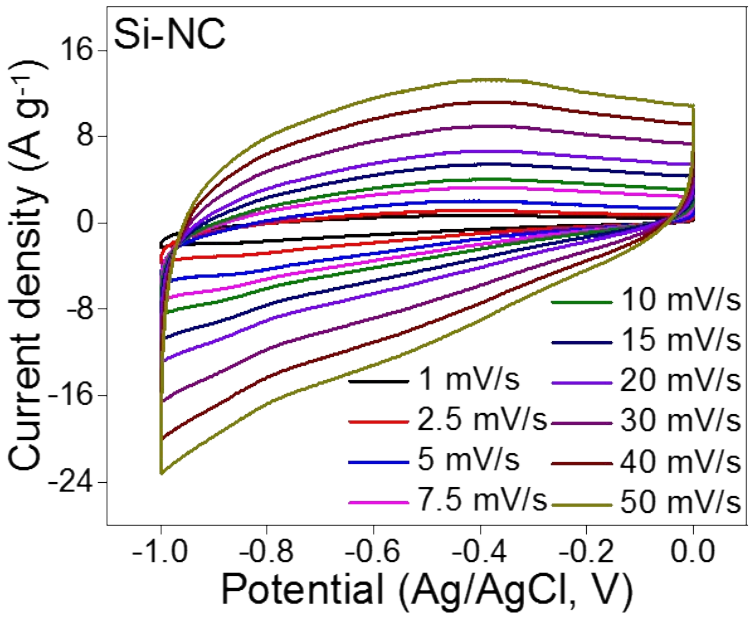

Figure S5: CV profiles of Si-NC electrode at various scan rates from 1 to 50 mV/s.

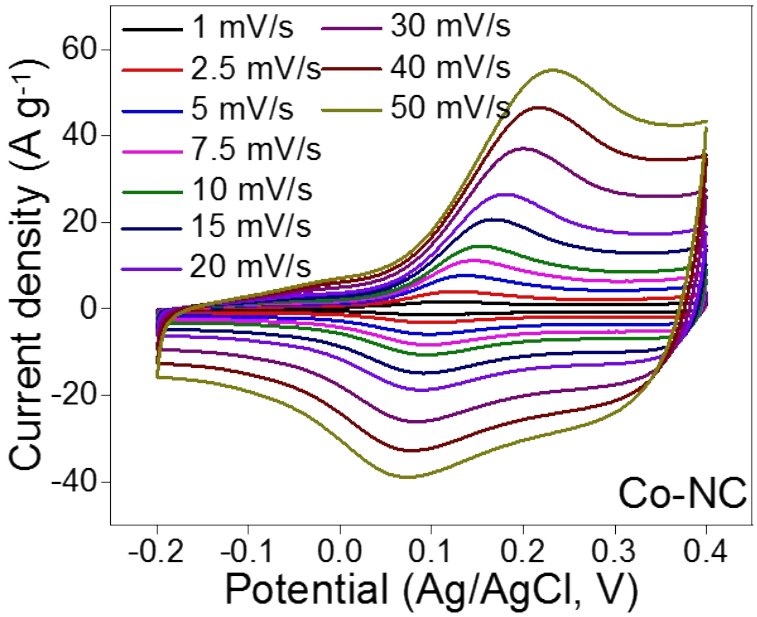

Figure S6: CV profiles of Co-NC electrode at various scan rates from 1 to 50 mV/s.

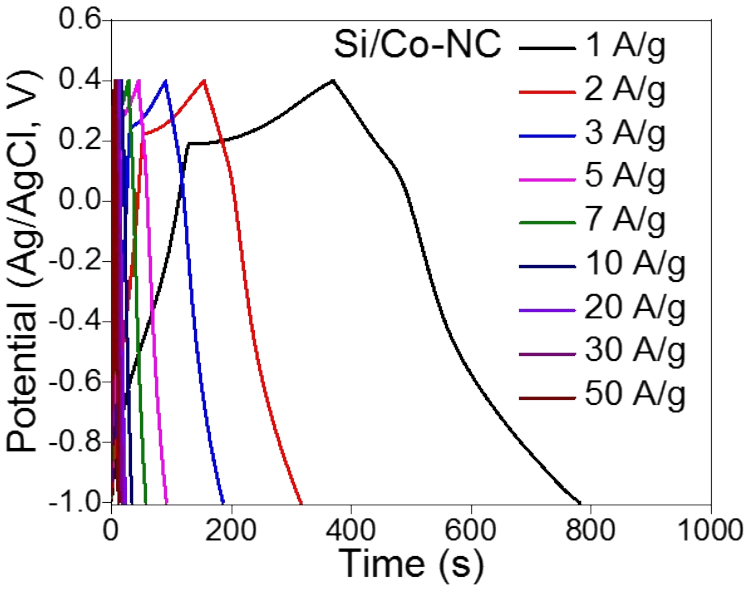

Figure S7: GCD profiles of Si/Co-NC integrated electrode at various current densities from 1 to 50 A/g.

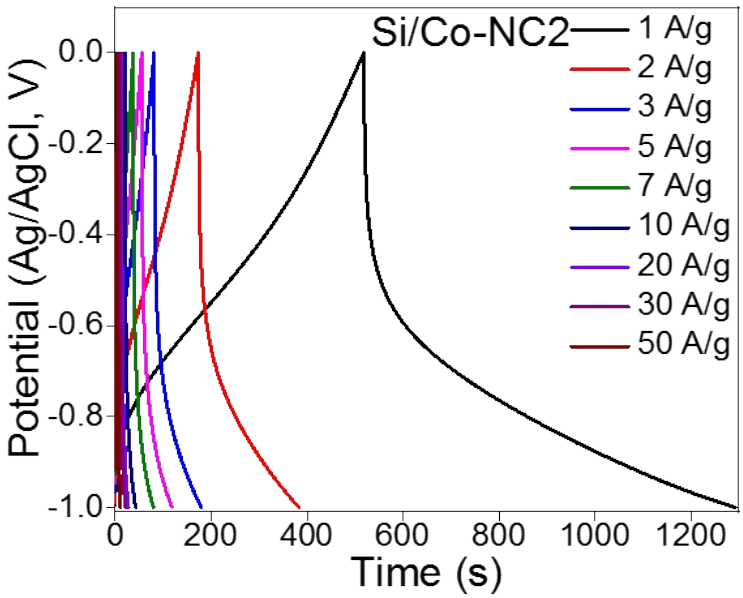

Figure S8: GCD profiles of Si/Co-NC2 integrated electrode at various current densities from 1 to 50 A/g.

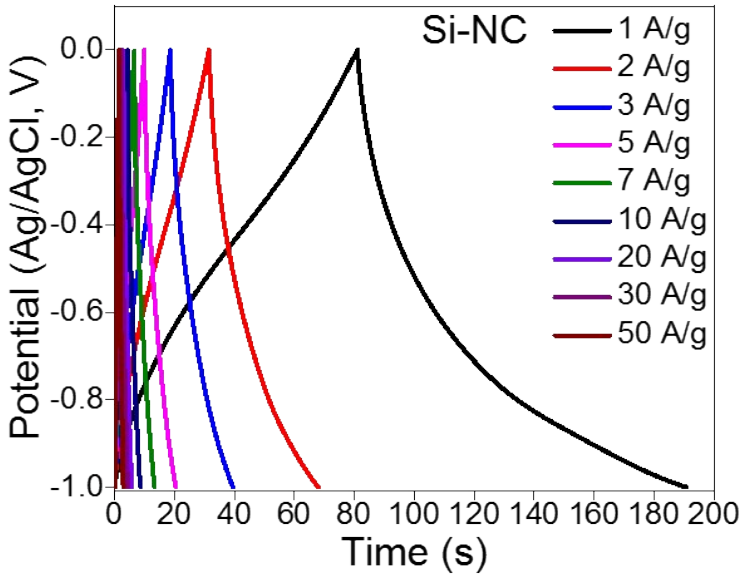

Figure S9: GCD profiles of Si-NC electrode at various current densities from 1 to 50 A/g.

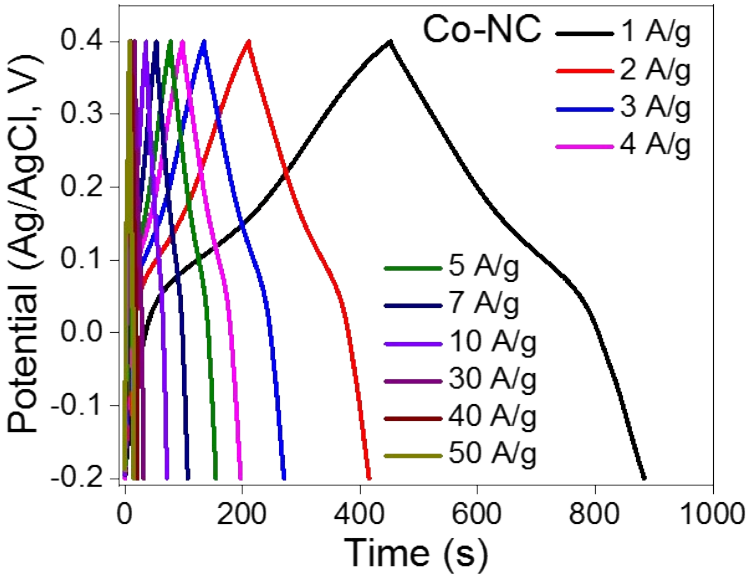

Figure S10: GCD profiles of Co-NC integrated electrode at various current densities from 1 to 50 A/g.

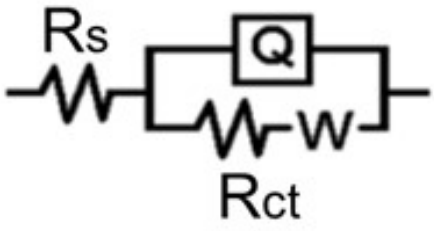

Figure S11: The equivalent circuit diagram for the analysis of EIS plot.

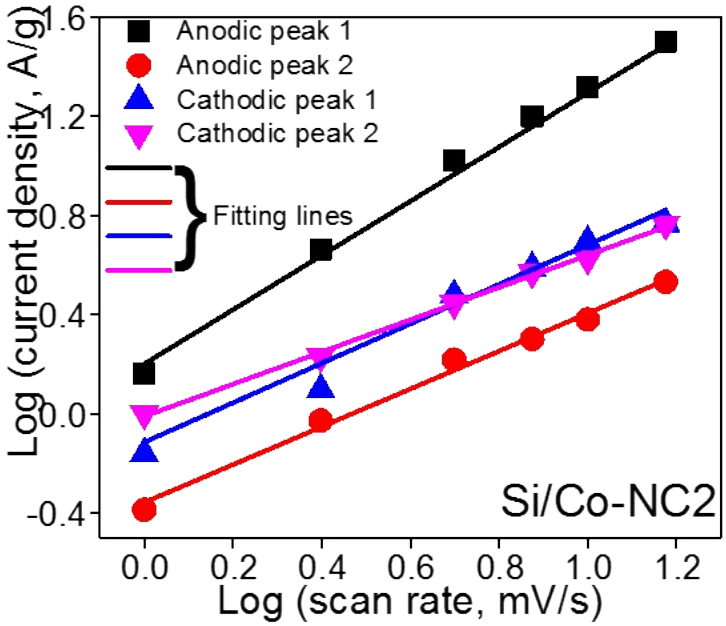

Figure S12: *b*-values calculated from log (*i*) versus log (*v*) of Si/Co-NC2.

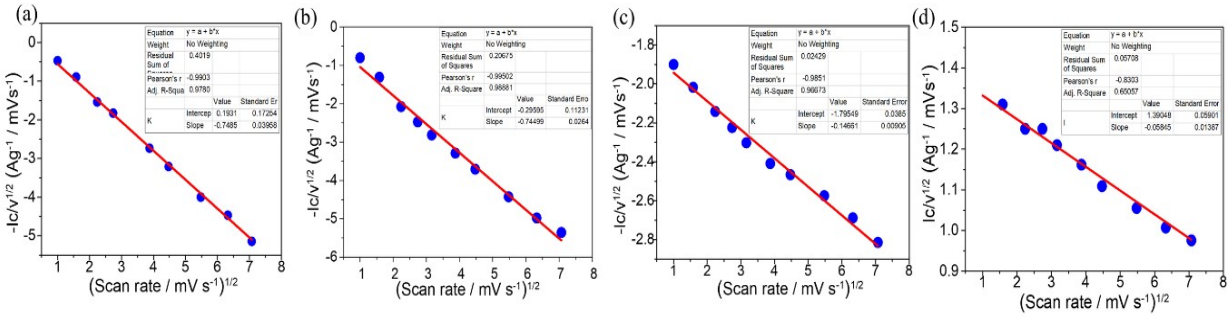

Figure S13: The calculation of k_1 and k_2-values in KOH aqueous electrolyte at various scan rates for (a) Si/Co-NC, (b) Si/Co-NC1, (c) Si/Co-NC2, and (d) Co-NC

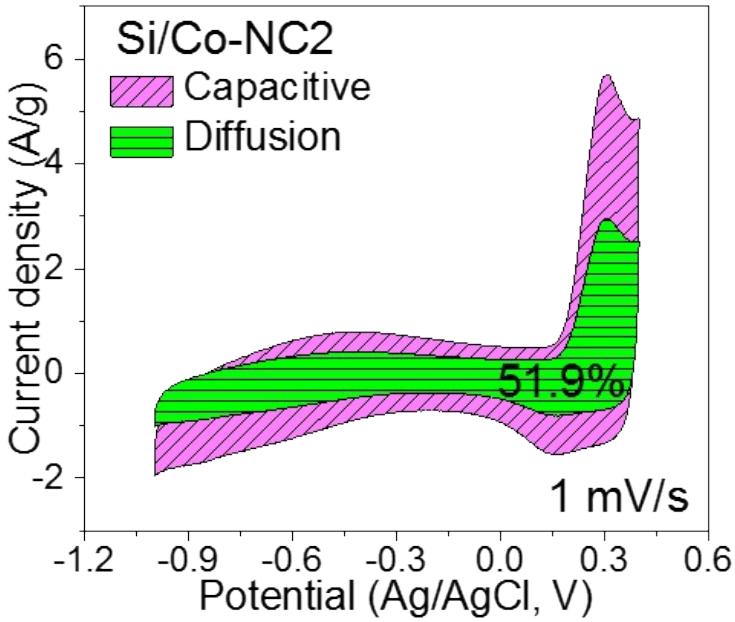

Figure S14: Percentage capacitive-controlled contribution in total charge storage process of Si/Co-NC2.

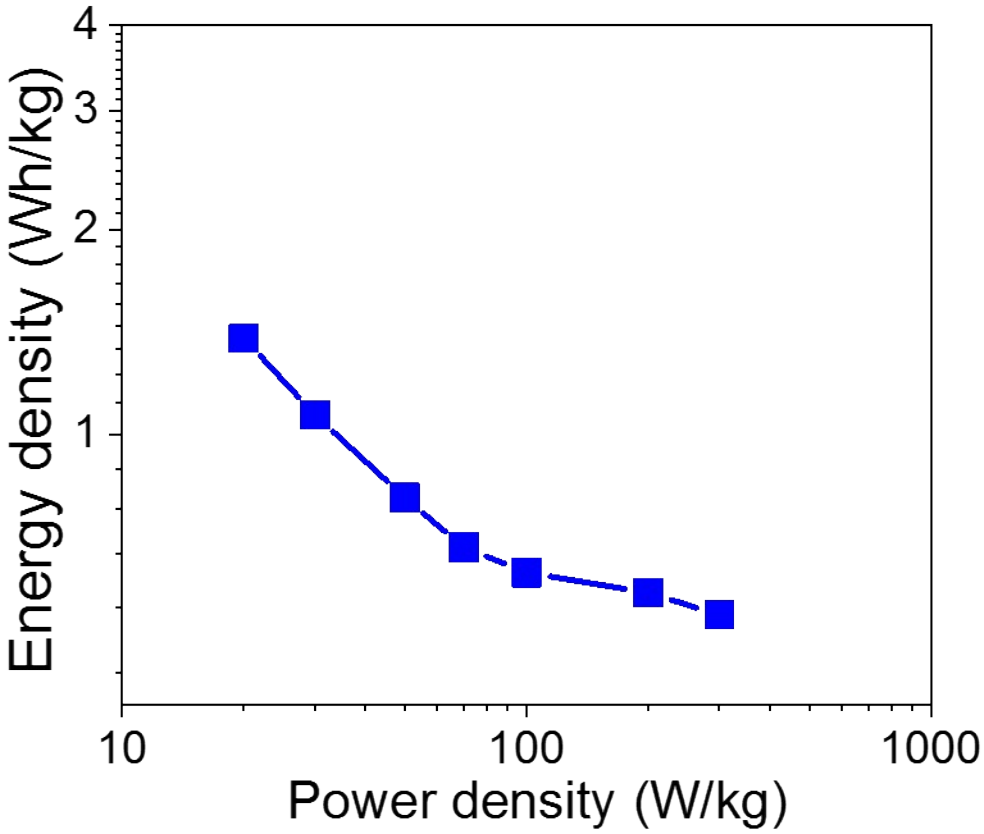

Figure S15: Ragone plot of Si/Co-NC1||Si/Co-NC1-SPC using the SC mass (containing electrodes/separator/electrolyte).

Table S3: A comparison table of electrochemical performances of bimetallic oxides and integrated system based three and two electrode supercapacitors.

Material three-electrode	**Electrolyte**	**Capacitance in 3-electrode (F/g) at 1A/g**	**Rate/ Cycles**	**Material two-electrode**	**Capacitance in 2 electrode (F/g) at 2 A/g**	**Rate/Cycles**	**E (Wh/kg) / P (W/kg)**	**Ref**
Si/Co-NC1	KOH	850	97%/ 10,000	Si/Co-NC1// Si/Co-NC1	276	96.8%/ 50,000	125/3500	This work
ZFO-ACFs	KOH	192	92.7%/ 20,000	ZFO-ACFs// ZFO-ACFs	45	-	27.6/523.6	[1]
$CoFe_2O_4$/graphene/PANI	KOH	767.7	---		392.3	96%/5000	79.7/178. 2	[2]
$NiCo_2O_4$-CNT@DNA	KOH	760	96. 2/5000	$NiCo_2O_4$-CNT@DNA//AC	223.7	90.3%/ 5000	69.7/373.9	[3]
$NiCo_2O_4$-UNSA	KOH	7.29	88.5%/5000	$NiCo_2O_4$-UNSA@$NiMoO_4$//AC	148	-	52.6/332.4	[4]
NiNTAs@Fe_2O_3	Na_2So_4	418.7	93.3%/ 5000	NiNTAs@Fe_2O_3//NiNTAs@MnO_2	95.9	92.3%/ 5000	34.1/3197.7	[5]
$ZnCo_2O_4$	KOH	776.2	84.3%/5000	$ZnCo_2O_4$// RGO	66	-	84.84/400	[6]
$NiCo_2O_4$	KOH	200	62.5%/ 5000	$NiCo_2O_4$// GO	61	-	38.53/299.3	[7]

References:

1. S. Yang, Z. Han, F. Zheng, J. Sun, Z. Qiao, X. Yang, L. Li, C. Li, X. Song and B. Cao, *Carbon*, 2018, **134**, 15-21.
2. P. Xiong, H. Huang and X. Wang, *Journal of Power Sources*, 2014, **245**, 937-946.
3. Y. Xue, T. Chen, S. Song, P. Kim and J. Bae, *Nano Energy*, 2019, **56**, 751-758.
4. P. Zhang, J. Zhou, W. Chen, Y. Zhao, X. Mu, Z. Zhang, X. Pan and E. Xie, *Chemical Engineering Journal*, 2017, **307**, 687-695.
5. Y. Li, J. Xu, T. Feng, Q. Yao, J. Xie and H. J. A. F. M. Xia, *Advanced Functional Materials*, 2017, **27**, 1606728.
6. L. Xu, Y. Zhao, J. Lian, Y. Xu, J. Bao, J. Qiu, L. Xu, H. Xu, M. Hua and H. Li, *Energy*, 2017, **123**, 296-304.
7. Y. V. Kaneti, R. R. Salunkhe, N. L. Wulan Septiani, C. Young, X. Jiang, Y.-B. He, Y.-M. Kang, Y. Sugahara and Y. Yamauchi, *Journal of Materials Chemistry A*, 2018, **6**, 5971-5983.