Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

Disclosing the Superior Lithium Storage of Double-Shelled Si@N-Doped Carbon: A Synergic Combination of Experiment and Theory

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Figures S1 to S9, Tables S1.

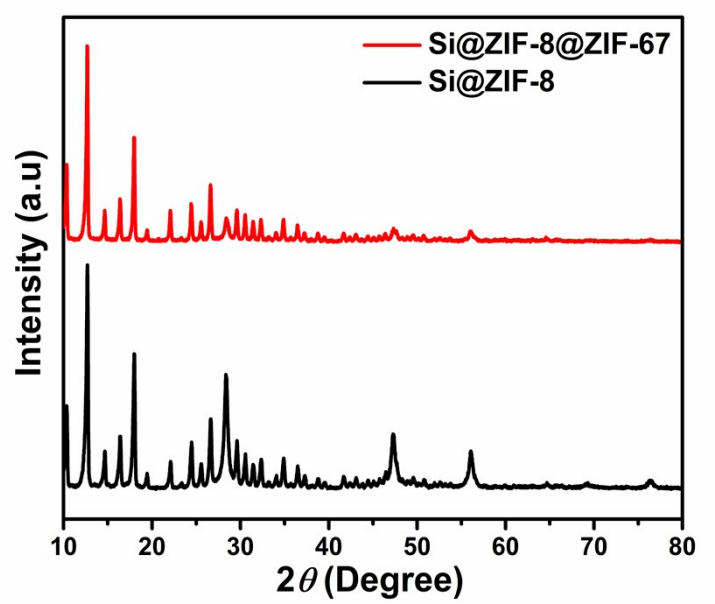

Figure S1: XRD pattern of single-shelled Si@ZIF-8 and double-shelled Si@ZIF-8@ZIF-67.

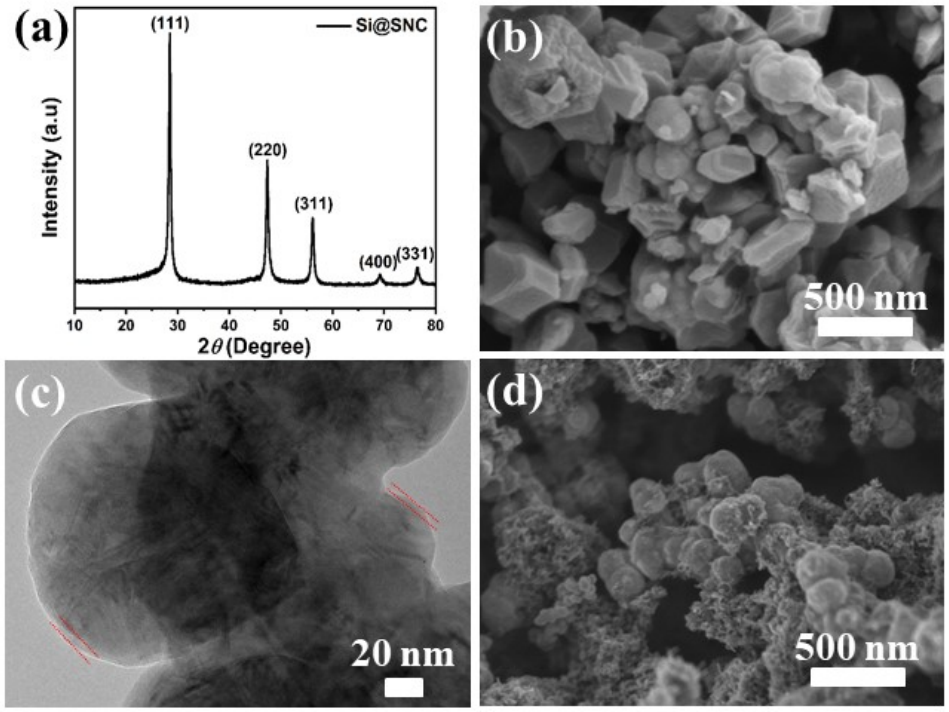

Figure S2: (a) XRD pattern of Si@SNC, (b) SEM image of Si@ZIF-8, (c) TEM image, and (d) SEM image of Si@SNC.

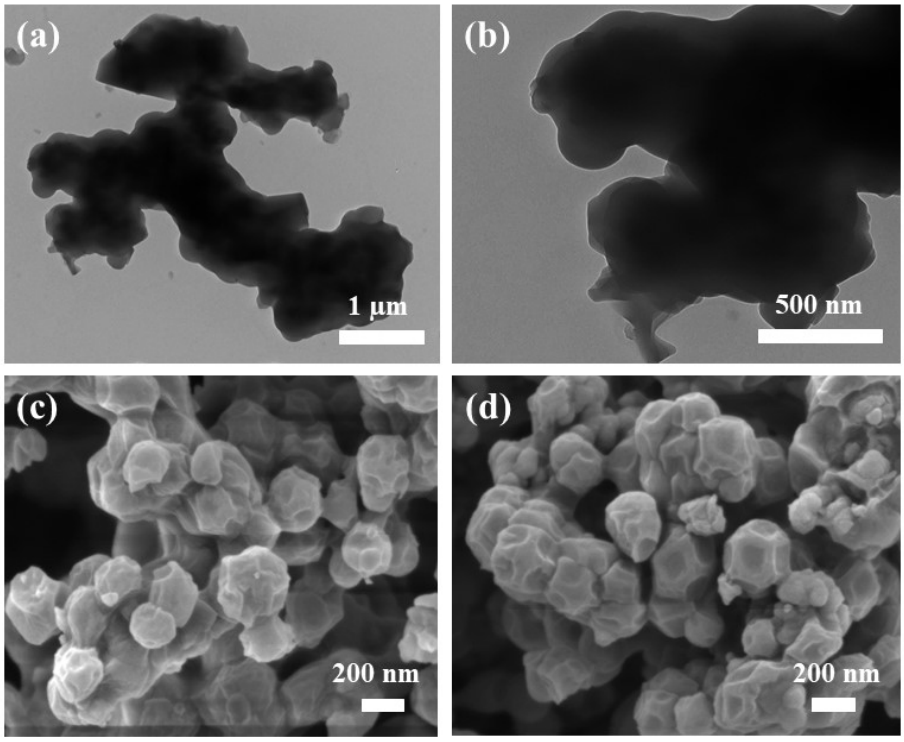

Figure S3: (a, b) TEM and (c, d) SEM images of Si@ZIF-8@ZIF-67.

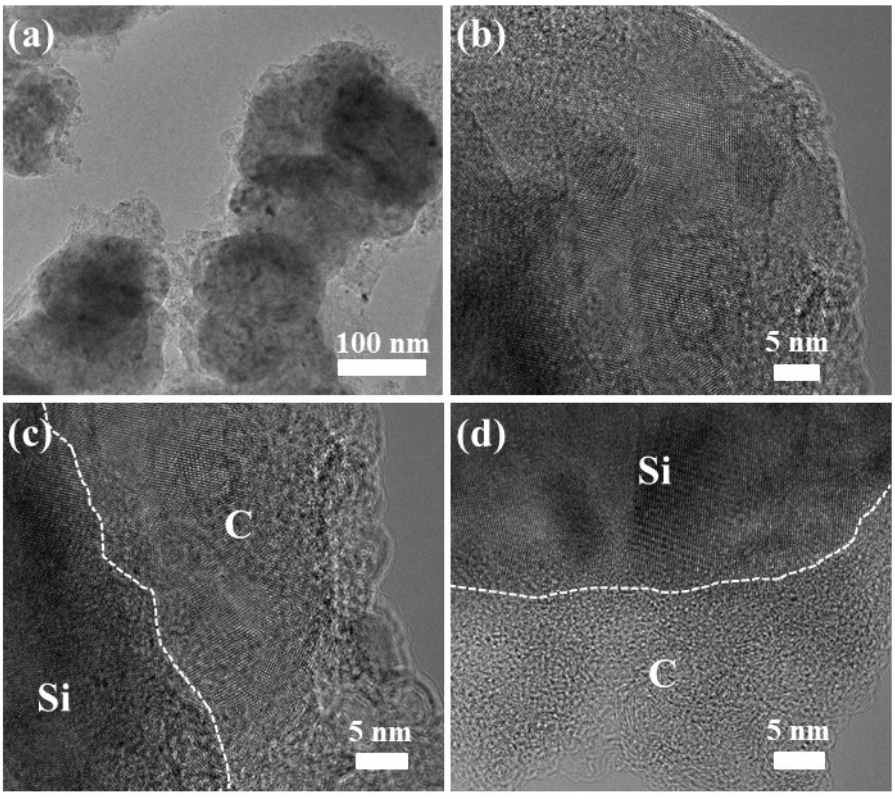

Figure S4: HRTEM images of double-shelled Si@DNC.

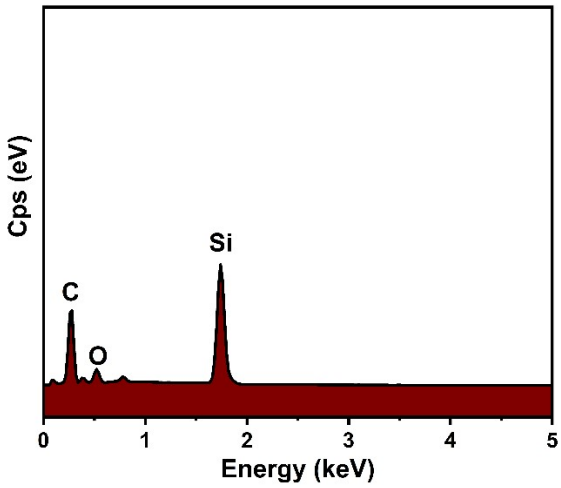

Figure S5: EDS spectrum of double-shelled Si@DNC.

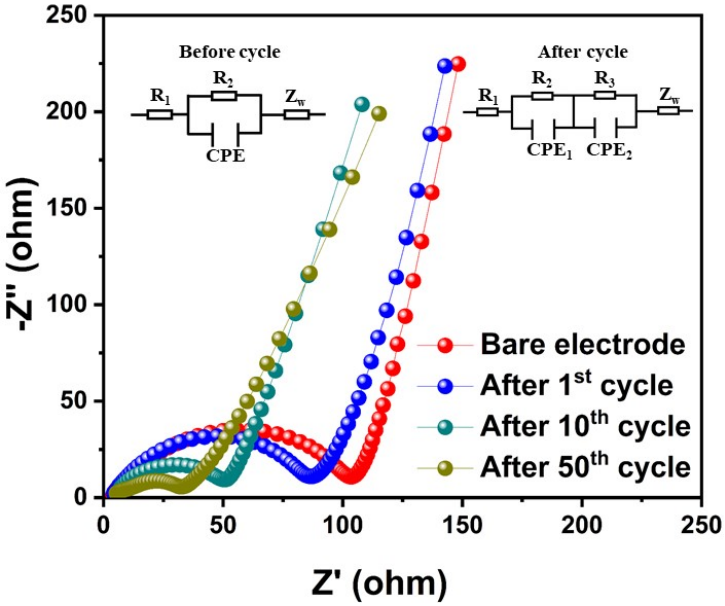

Figure S6: Electrochemical impedance spectra of Si@DNC for bare electrode, after 1st cycle, 10th cycles, and 50th cycles.

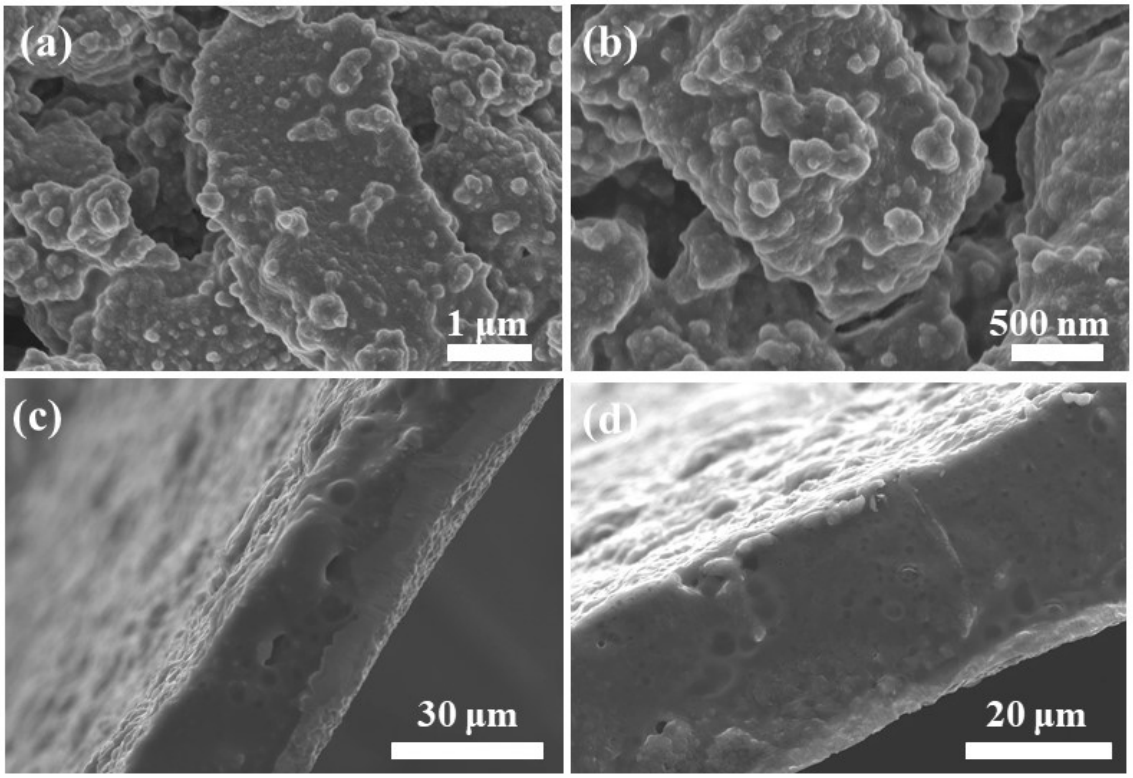

Figure S7: (a-d) SEM images of double-shelled Si@DNC cycled electrode.

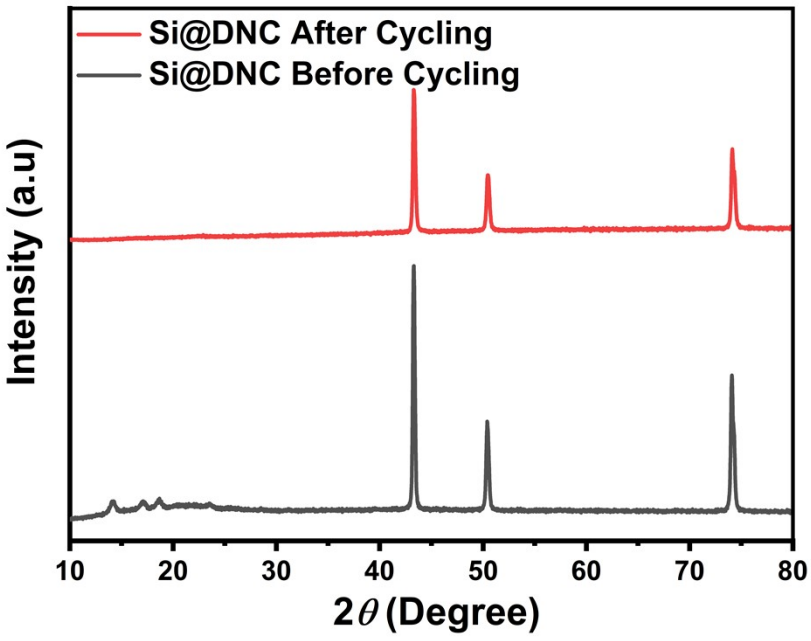

Figure S8: XRD patterns of Si@DNC electrode before and after 300 cycles.

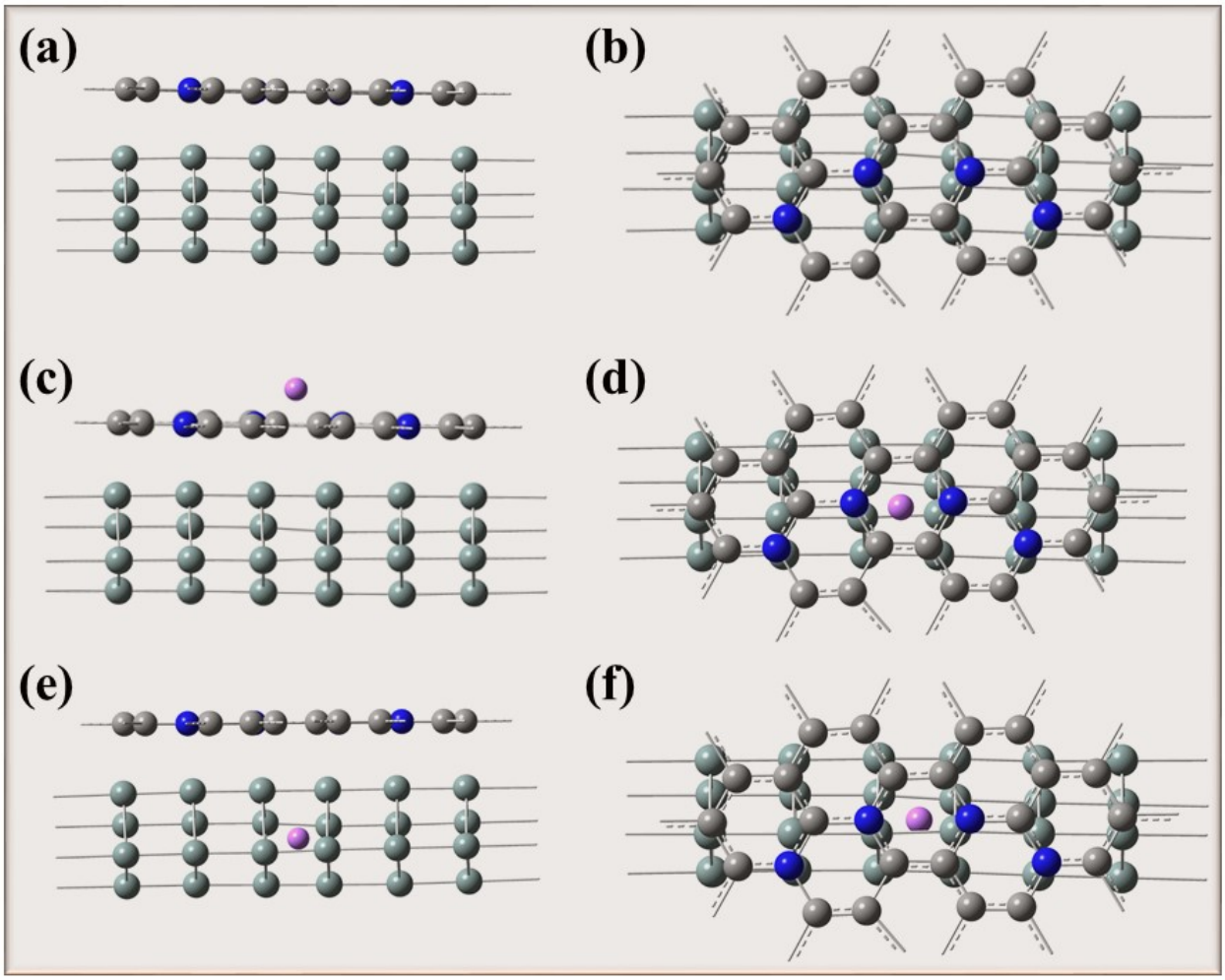

Figure S9: (a-c) side view and (d-f) top view of optimized lithiation mechanism of single shelled Si@SNC.

Table S1. Calculated quantum molecular descriptors (eV) for optimized lithiation mechanism.

Materials	HOMO	LUMO	E (RB3LYP)	B.E.	E_{HOMO}	E_{LUMO}	Eg	I=- E_{HOMO}	A=- E_{LUMO}	μ (Debye)	η	ω
N-carbon	0.20	-0.11	-628.88	-	-5.46	-312	2.34	5.46	3.12	-4.29	1.17	7.87
N, N-carbon	-0.11	-0.06	-645.37	-	-3.07	-1.65	1.42	3.07	1.65	-2.36	0.71	3.92
Si	-0.21	-0.12	-2879.70	-	-5.85	-3.44	2.41	5.85	3.44	-4.64	1.20	8.95
Li	-1.90	-0.05	-7.45	-	-51.87	-1.41	50.46	51.87	1.41	-26.65	25.23	14.06
N-N-carbon-Si	-0.20	-0.11	-3674.83	-8.69	-5.50	-3.12	2.38	5.50	3.12	-4.31	1.19	7.81
Li-N-N-carbon-Si	-0.17	-0.15	-3682.39	-11.36	-4.67	-4.13	0.55	4.67	4.13	-4.40	0.27	35.85
N, N-carbon-Si-Li	-0.17	-0.16	-3682.41	-11.94	-4.61	-4.30	0.31	4.61	4.30	-4.46	0.16	64.02
N-carbon-N-carbon-Si	-0.16	-0.15	-4303.71	-8.61	-4.28	-3.97	0.32	4.28	3.97	-4.13	0.16	54.89
Li-carbon-carbon-Si	-0.15	-0.14	-4311.12	-7.14	-4.11	-3.88	0.23	4.11	3.88	-4.00	0.12	69.39
Carbon-Li-carbon-Si	0.16	-0.15	-4310.99	-3.90	-4.25	-4.21	0.04	4.25	4.21	-4.23	0.02	447.32
Carbon-Carbon-Si-Li	-0.17	-0.17	-4311.05	-3.31	-4.74	-4.66	0.08	4.74	4.66	-4.70	0.04	276.13