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Electronic Supplementary Information

Cost-effective approach for structural evolution of Si-based multicomponent for Li-ion battery anodes

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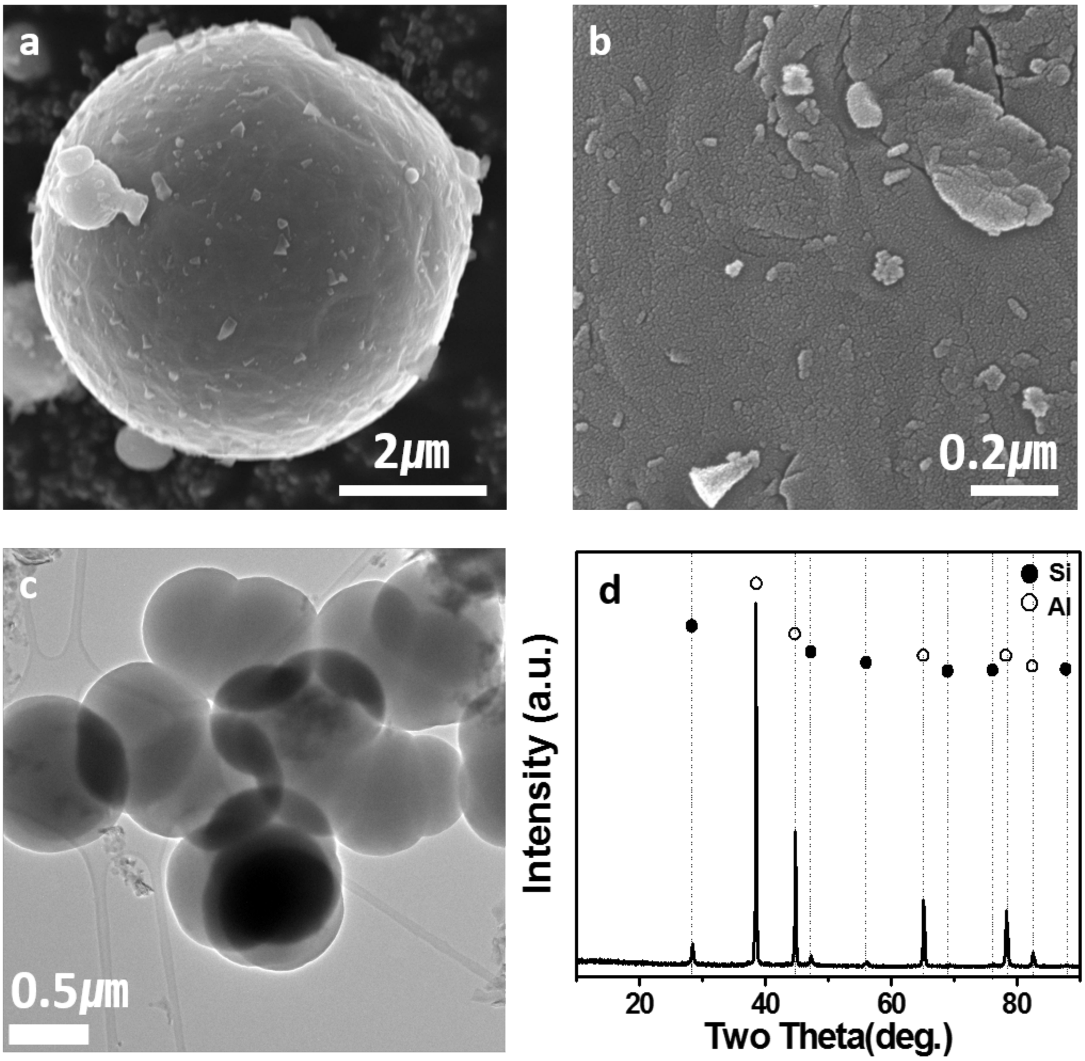
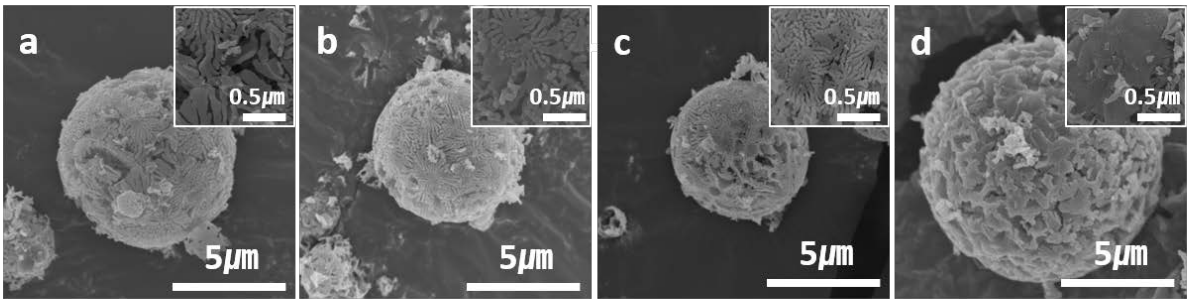


Figure S1. Characterization of pristine Al-Si alloy. (a-b) SEM images, (c) TEM image and (d) XRD pattern of pristine Al-Si alloy



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(wt%)	Pristine	Al-Si-4	Al-Si-10	Al-Si-20	Al-Si-40
O	4.45	7.56	7.27	7.45	8.04
Al	84.69	4.28	10.38	21.10	39.65
Si	10.87	88.16	82.34	71.45	52.30

Figure S2. Structural analysis of etched Al-Si. SEM images of etched Al-Si (a) 04, (b) 10, (c) 20 and (d) 40 samples. (e) EDX quantification results of each samples.

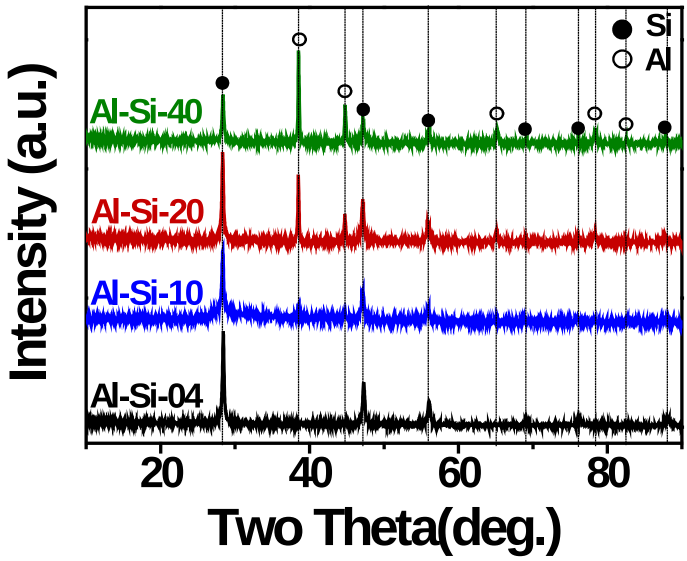


Figure S3. Crystal structure analysis of etched Al-Si. XRD pattern of Etched Al-Si.

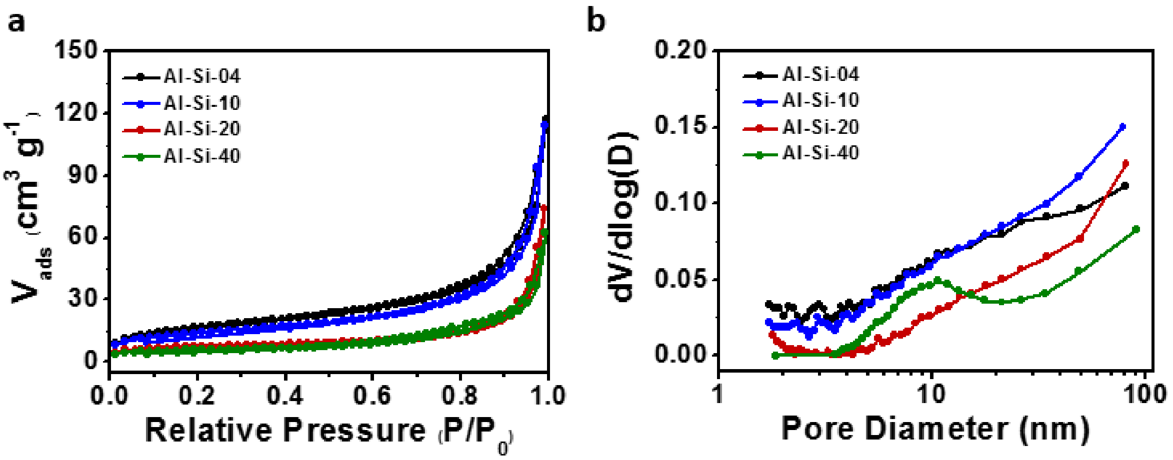


Figure S4. Porosity characterization of etched Al-Si. (a) Nitrogen adsorption isotherm and (b) BJH pore size distribution of etched Al-Si.

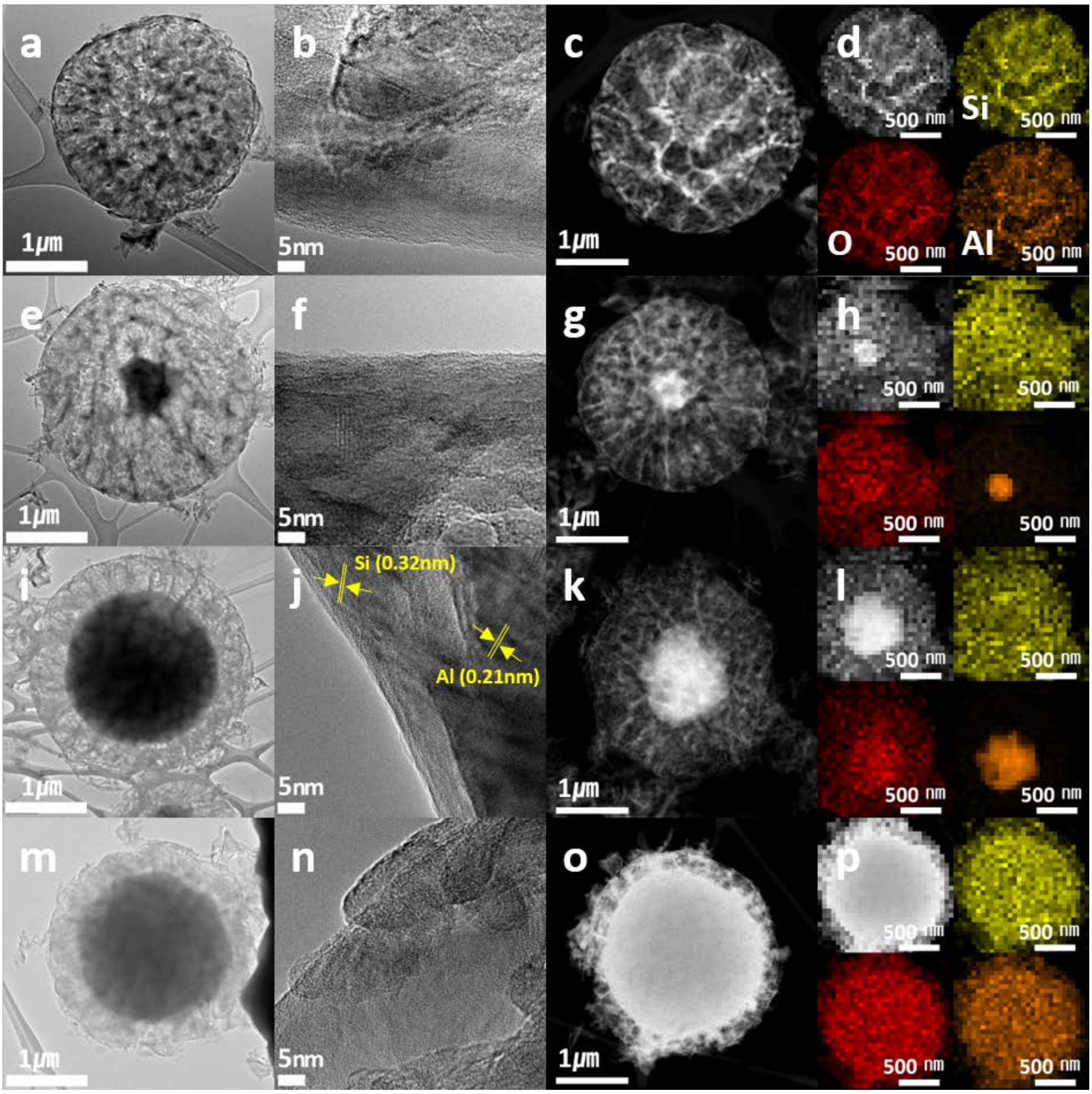
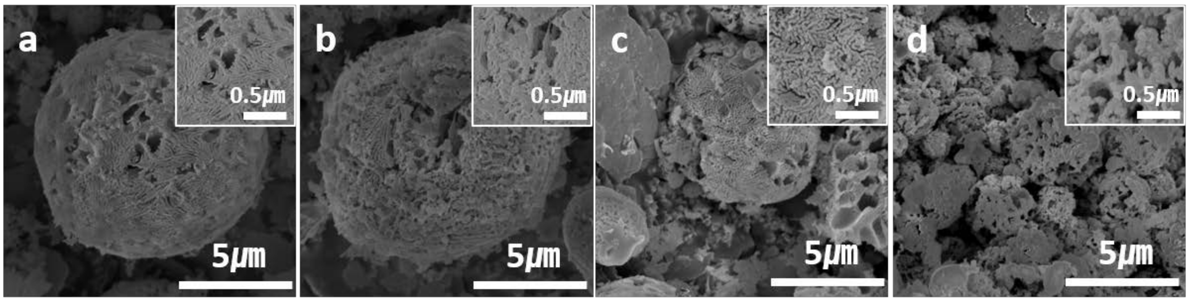


Figure S5. TEM characterization of etched Al-Si. TEM images, STEM-HAADF and EDX mapping results of etched Al-Si (a-d) 04, (e-h) 10, (i-l) 20 and (m-p) 40 samples.



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(wt%)	ASWO-4	ASWO-10	ASWO-20	ASWO-40
O	11.85	13.57	16.24	20.63
Al	2.82	8.71	14.69	30.37
Si	85.32	77.73	69.07	49.00

f

	Al-4wt%		Al-10wt%		Al-20wt%		Al-40wt%	
	Etched	Oxidized	Etched	Oxidized	Etched	Oxidized	Etched	Oxidized
Si/Al	20.60	30.26	7.93	8.92	3.39	4.70	1.32	1.61
	46.9% increase		12.5%		38.6%		22.0%	

Figure S6. Structural analysis of wet-oxidized etched Al-Si (ASWO). SEM images of ASWO (a) 04, (b) 10, (c) 20 and (d) 40 sample. (e-f) EDX quantification results.

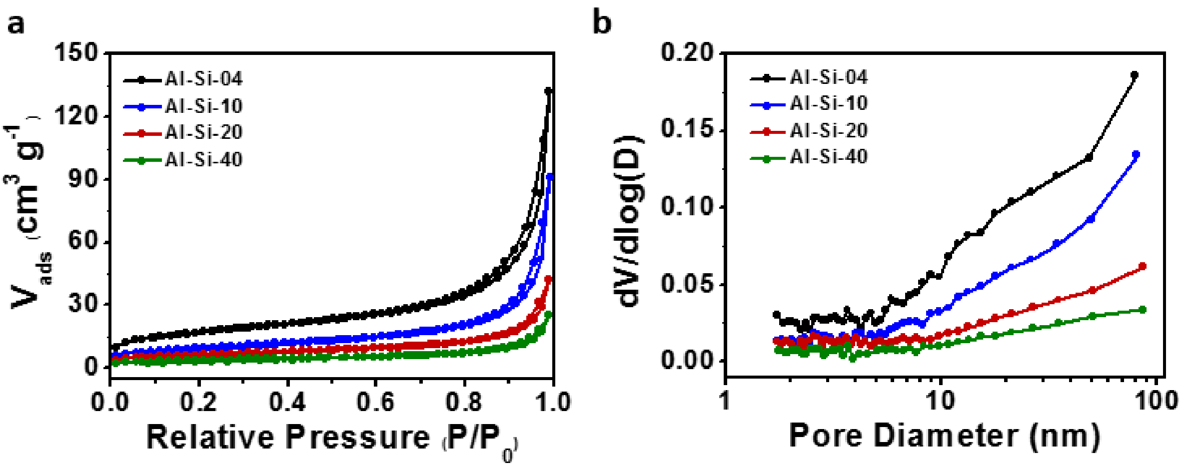
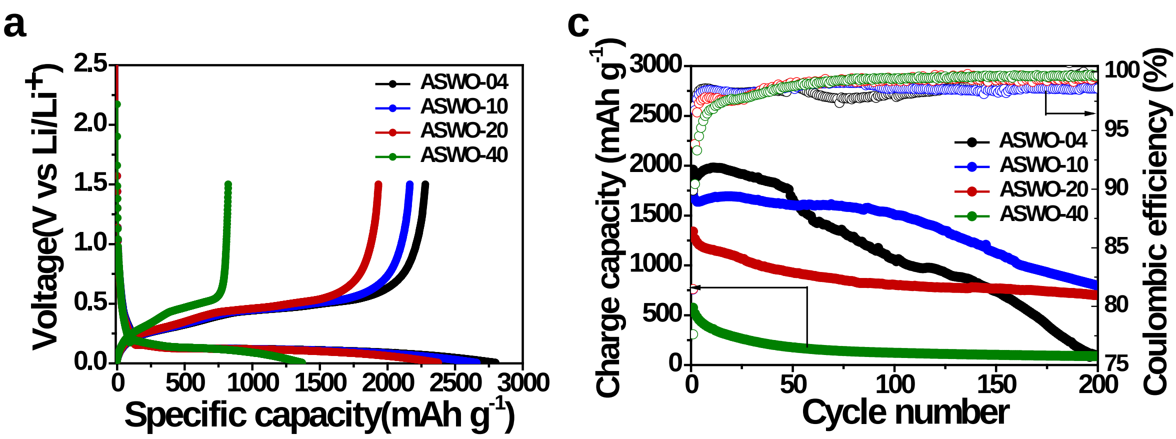


Figure S7. Porosity characterization of ASWO. (a) Nitrogen adsorption isotherm and (b) BJH pore size distribution of ASWO samples.



b

	ASWO-4	ASWO-10	ASWO-20	ASWO-40
C_{ch} (mAh g^{-1})	2279.6	2165.6	1933.6	822.6
ICE (%)	81.4	81.4	80.5	60.1

Figure S8. **Electrochemical properties of etched Al-Si in half cell.** (a) Initial charge-discharge profiles at C/20, (b) Summary chart for capacity and ICE, and (c) Cyclic performances for 200 cycles at C/5.

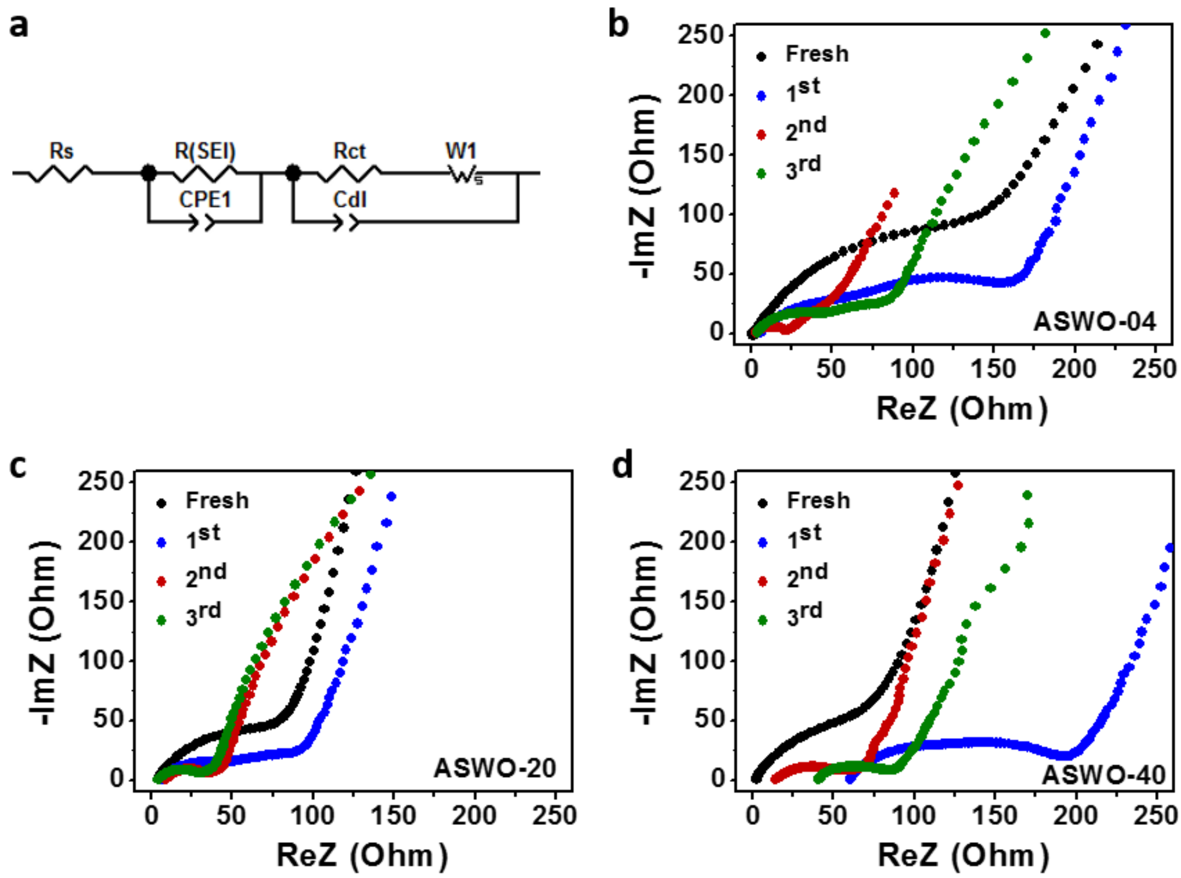


Figure S9. **Electrochemical Impedance spectroscopy (EIS) analysis of ASWO electrodes.**

(a) Equivalent Short circuit model of electrochemical impedance spectroscopy measurement. Ex-situ impedance analysis of (b) ASWO-04, (c) ASWO-20 and (d) ASWO-40 electrodes.

	4wt%	10wt%	20wt%	40wt%
Pristine (μm)	10.5	17.5	19.5	10.1
Lithiation (μm)	19.5	20.2	26.8	19.1
After 30cycle (μm)	16.1	20.7	27.1	18.7
Volume change (%) Pristine → Litiation	**85.7**	**15.4**	**37.4**	**89.1**
Volume change (%) Pristine → After cycle	**53.3**	**18.2**	**39.0**	**85.1**

Figure S10. Summary chart for electrode swelling results.

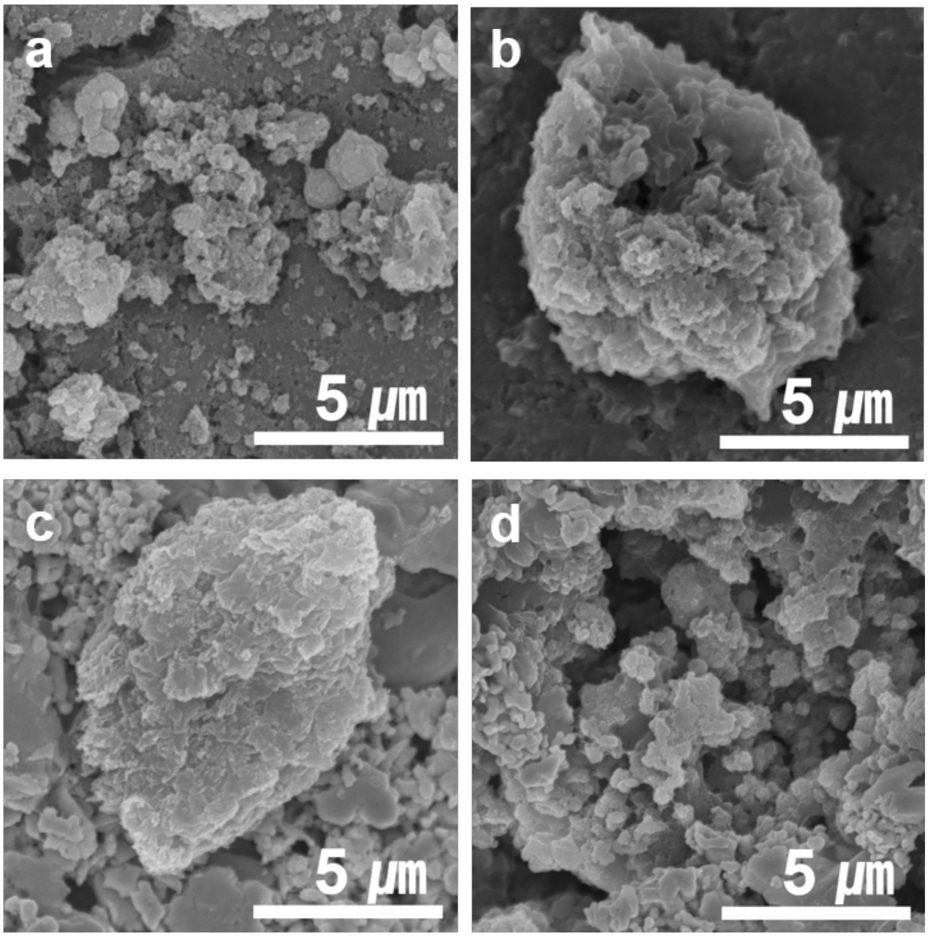
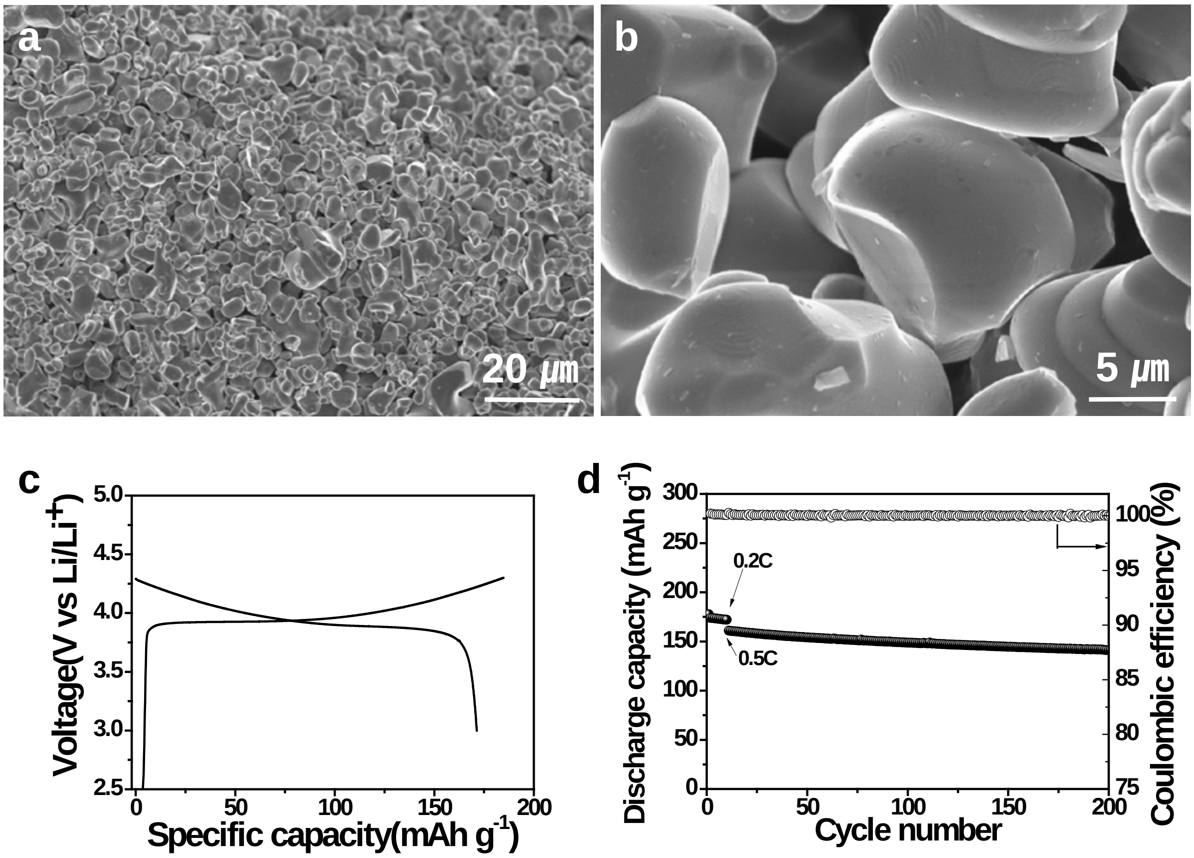


Figure S11. Morphological changes of ASWO samples after 30 cycles at a rate of 0.2 C. SEM images of (a) ASWO-04, (b) ASWO-10, (c) ASWO-20 and (d) ASWO-40 samples.



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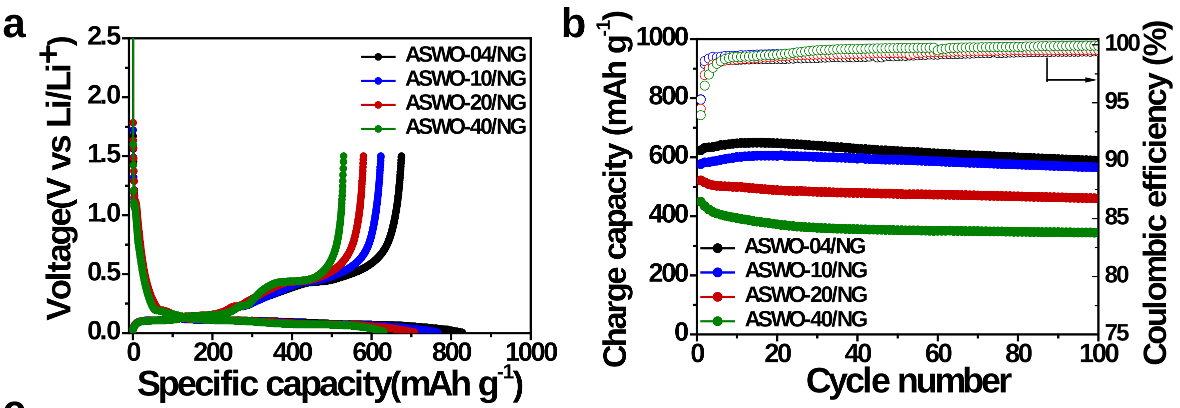
	Initial C_{ch} (mAh g^{-1})	Initial C_{dis} (mAh g^{-1})	ICE(%)	Capacity retention (%) after 200 cycles
$LiCoO_2$	184.7	171.3	92.74	87.4

Figure S12. Properties of $LiCoO_2$ (LCO) cathode in half cell. (a-b) SEM images, (c) initial charge-discharge profiles at a rate of 0.1 C, (d) cyclic performance for 200 cycles at a rate of 0.5 C and (e) summarized electrochemical properties of LCO cathode.



c

	ASWO-4	ASWO-10	ASWO-20	ASWO-40
C_{dis} (mAh g^{-1})	675.1	623.5	579.9	529.7
ICE (%)	81.6	81.3	81.8	81.3
Capacity retention (%) After 100 cycles	94.4	98.2	88.3	76.6

Figure S13. Electrochemical properties of ASWO/NG anode in half cell. (a) Initial charge-discharge profiles at a rate of 0.05 C, (b) cyclic performance for 100 cycles at a rate of 0.2 C and (c) summarized electrochemical properties of ASWO/NG electrodes.

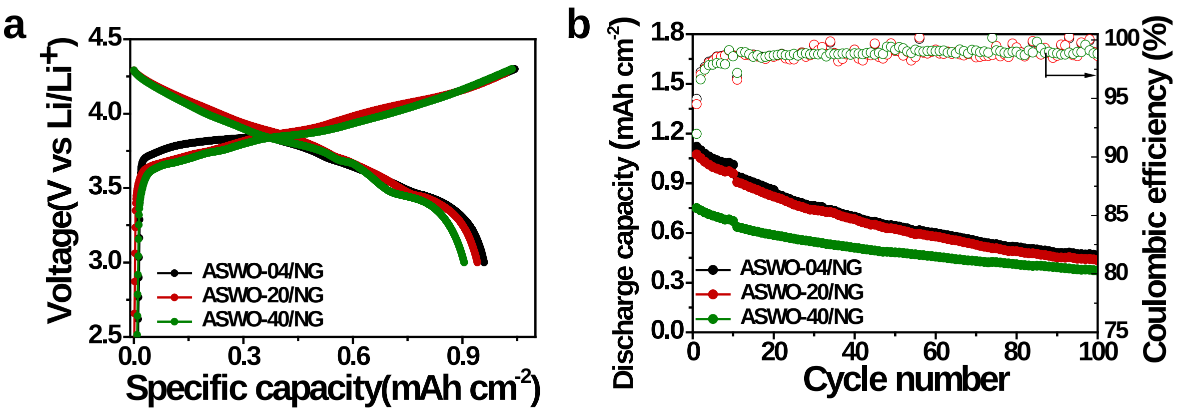


Figure S14. Electrochemical properties of ASWO/NG-LCO full cell. (a) Initial charge-discharge profiles at a rate of 0.1 C and (b) cyclic performance for 200 cycles at a rate of 0.5 C of NG/ASWO 04/20/40 electrodes.

Table S1. Summary for electrochemical test conditions.

	Slurry loading	Electrode composition	Active materials	Active materials loading	Gravimetric capacity		Areal capacity	Current density	
					Slurry	Active		1C	1C
	mg cm^{-2}	*AM : C : B	(Si %)	mg cm^{-2}	mAh g^{-1}	mAh g^{-1}	mAh cm^{-2}	mA g^{-1}	mA cm^{-2}
Al-Si-04	1.94	70 : 10 : 20	Al, Si (61.7)	1.36	1595.7	2279.6	3.10	2279.6	3.10
Al-Si-10	1.67	70 : 10 : 20	Al, Si (57.6)	1.17	1515.9	2165.6	2.53	2165.6	2.53
Al-Si-20	1.76	70 : 10 : 20	Al, Si (50.0)	1.23	1353.5	1933.6	2.38	1933.6	2.38
Al-Si-40	1.67	70 : 10 : 20	Al, Si (36.6)	1.17	575.8	822.6	0.96	822.6	0.96
ASWO-04	1.49	70 : 10 : 20	Al_2O_3, Si (59.2)	1.04	994.0	1420.0	1.48	1420.0	1.48
ASWO-10	1.67	70 : 10 : 20	Al_2O_3, Si (54.4)	1.17	895.0	1278.6	1.50	1278.6	1.50
ASWO-20	2.03	70 : 10 : 20	Al_2O_3, Si (48.3)	1.42	820.3	1171.8	1.75	1171.8	1.75
ASWO-40	2.13	70 : 10 : 20	Al_2O_3, Si (34.3)	1.49	506.8	724.0	1.17	724.0	1.17
LCO	5.56	85 : 7.5 : 7.5	LCO (-)	3.89	119.9	171.3	0.67	171.3	0.67
ASWO-10/NG	2.13	90 : 2 : 8 (ASWO : NG = 15 : 75)	Al_2O_3, Si, NG (12.3)	1.49	436.5	623.5	0.93	623.5	0.93

* AM = Active Materials
C = Conducting carbon
B = Binder

Table S2. Summary for battery performance of various Si-based multicomponent system.

Si anode	Potential window (V vs. Li/Li^+)	Anode half cell							Full cell					Ref.
		First cycle			Capacity after X cycles at Y C-rate				Cathode	First cycle	Capacity retention after x cycles at Y C-rate			
		1^{st} C_{dis} ($mAh\ g^{-1}$)	1^{st} C_{ch} ($mAh\ g^{-1}$)	ICE (%)	X	Y	Capacity / retention ($mAh\ g^{-1}$) / (%)			ICE (%)	X	Y	retention (%)	
Si/Al_2O_3	0.01-1	~2550	2055	~80	100	0.2	~1500	82	-	-	-	-	-	26
Al_2O_3 coated NiSi-SiNW	0.01-2	~3900	~3400	~87	100	0.2	~2600	~75	-	-	-	-	-	27
Si-Al_2O_3	0.23-2	-	1125	-	100	-	~1100	~98	-	-	-	-	-	28
Si-$NiSi_2$-$Al_2O_3@C$	0.01-2	~1000	~800	~80	50	0.1	~700	~89	-	-	-	-	-	29
Si/hard carbon	0.005-1.5	654	513	78.4	100	0.2	300	~100	LMO	86.2	50	0.2	87	1*
Si/graphene	0.05-1	2782	2227	80.1	50	0.5	984	60.1	NCA	78.7	50	0.5	82.9	2*
Si@TiO2/C	0.005-2.5	1710	1260	74	50	0.2	950	90	LCO	69	-	-	-	3*
Si/graphene	0.01-1	1365	891	65.3	500	0.4	553	~65	LNMO	78.3	100	0.2	~55	4*
Si/CNT	0.005-3	-	~2240	-	100	0.4	~2000	88	CNT/ LMO	~88	100	1	87	5*
Our work (ASWO-10)	**0.005-15**	**1622.6**	**1278.6**	**78.8**	**500**	**0.2**	**~810**	**81.9**	**LCO**	**91**				**-**

***=supplementary references**

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