Supporting Information

Electrospray Synthesis of Nano-Si Encapsulated in Graphite/carbon Microplates as Robust Anode for High Performance Lithium-ion Batteries

Wen Liu ^a, Yongming Zhong ^a, Siyuan Yang ^a, Shengsen Zhang ^a, Xiaoyuan Yu ^a, Hongqiang Wang ^{b*}, Qingyu Li ^b, Jun Li ^c, Xin Cai ^{a*}, Yueping Fang ^a

^{*a*} College of Materials and Energy, South China Agricultural University, Guangzhou, Guangdong 510642, China.

^b Guangxi Key Laboratory of Low Carbon Energy Materials, School of Chemistry & Pharmaceutical Sciences of Guangxi Normal University, Guilin 541004, P. R. China.

^c Department of Chemistry, Kansas State University, Manhattan, KS 66506, United States.



Figure S1. (a), (b) SEM images of Si/G composite from ball milling. (c), (d) SEM images of Si/G/C composite. (e), (f) SEM images of Si/C through electrospray and pyrolysis.



Figure S2. HR-TEM image of Si/G/C. the area A and B in red dashed box indicates the crystalline zone and the amorphous zone of silicon particles, respectively.



Figure S3. Deconvoluted O 1s XPS spectra derived from Si/G/C.



Figure S4. (a) N_2 adsorption-desorption isotherms of Si/G/C at 77 K. (b) Barrett-Joyner-Halenda desorption pore-size distribution of Si/G/C.

Table S1 Comparison of the preparation and electrochemical performances of reported

 Si/graphite-based anode materials.

Active materials	Preparation method	Silicon	Current	Initial	Cycling performance	Ref.
		content	density	capacity		
		(wt%)	(mA/g)	(mAh/g)		
Nano-Si/Graphite composite	Ball milling;				800 mAh/g	
	polysaccharides	20	50	760~870	over 150	【1】
	binder				cycles	
Si@SiO _x @Ni/Gra phite composite	Two-step ball milling from SiO	31.8	1000	1444.7	601 mAh/g	
					over 50	[2]
					cycles	
Graphite/Si@C	Mechanical milling,				570.7 mAh/g	
	spray drying and pitch	15.7	~100	637.7	after 100	[3]
	pyrolysis				cycles	
Ci ananhita hindan					514 mAh/g	
Si-graphite-binder (polyetherimide)	Ball milling	5	~50	~-1550	after 350	【4】
					cycles	
Si/C microrods on					590.5 mAh/g	
graphite	CVD process	12	50	616.7	after 50	[5]
microspheres					cycles	
Ultranano- Si/Graphite flakes	Etching of recycled Si wafer and ball milling	75	800	~1500	1200 mAh/g	
					over	[6]
					300 cycles	
Nano-	Doll milling and					
Si/Graphite/C spherical composite	petroleum pitch	20		~700	~650 over 50	【7】
					cycles	
	pyrolysis at 1000 C					
Si/C/Graphite composite	Carbonization of coal- tar pitch	17	130	712	569.6 mAh/g	
					after 100	[8]
					cycles	

	Thermal				850 m A h / ~	
Si/Graphite	decomposition of	21 (040	1	850 mAn/g	[0]
composite	cyclopentasilane	21.0	840	/	over 100	[9]
	precursor				cycles	
Si-nanolayer-					~500 mAh/g	
embedded	CVD process	6	~55	517	after 100	【10】
graphite/C hybrid					cycles	
Si-metal alloys/Graphite	Arc melting and ball	~7	94.8	~460	~400 mAh/g	
	milling; poly (vinyl				after 100	【11】
	alcohol) binders				cycles	
Nano-Si@C	Carbonization of BAN		50	757	680 mAh/g	
		9.8			after 100	[12]
	at 1000 C				cycles	
Nano	Ball milling, spray				400 mAh/s	
Nano-	drying, pyrolysis of	16	200	756	400 mAn/g	【13】
Si@Graphite/C	glucose and	16			after 300	
microspheres	polyvinylpyrrolidone				cycles	
Si/Flake	Ball milling and				517 mAh/g	
Graphite/Carbon	carbonization of	~17	~180	634	after 200	【14】
Composite	mesophase pitch				cycles	
	Ball milling and high-	17.3	168	~840	420 A h /	【15】
Si/Graphite/Disor dered carbon	temperature				420 mAn/g	
	annealing; different				after 260	
	electrolytes				cycles	
Nano-	Come da la cond				~500 mAh/g	
Si/graphite@grap hene composite	sintering	16%	100	803.3	after 50	[16]
					cycles	
Sub-micron					428 mAh/g	
silicon/Pyrolyzed	Spray-drying-assisted	6.7	100	556	after 100	【17】
carbon@graphite	self-assembly				cycles	
a: a a	TT' 1				610 mAh/g	
S1-C0-C	High energy	20	50	1283.3	after 50	【18】
composite	mechanical milling				cycles	
Nano-	Mixing; Using				~630 mAh/g	
silicon/graphite	conductive polymer	20	~100	~900	after 100	【19】
composite	binder				cycles	
Silicon/	High energy	70	237	1790	~700 mAh/g	
					after 25	[20]
Graphite-tin	mechanical milling				cycles	
Nano- Si/Graphite/C	Ball milling and	21.8	200	1295	523 mAh/g	This
					after 200	
microplates	electrospray				cycles	work
-					-	

References:

[1] M. Murase, N. Yabuuchi, Z. J. Han, J. Y. Son, Y. T. Cui, H. Oji, S. Komaba, Chemsuschem, 5 (2012) 2307-2311.

[2] J. Wang, W. Bao, L. Ma, G. Tan, Y. Su, S. Chen, F. Wu, J. Lu, K. Amine, Chemsuschem, 8 (2015) 4073-4080.

[3] J. Li, J. Wang, J. Yang, X. Ma, S. Lu, Journal of Alloys and Compounds, 688 (2016) 1072-1079.

[4] C. H. Yim, F.M. Courtel, Y. Abu-Lebdeh, Journal of Materials Chemistry A, 1 (2013) 8234-8243.

[5] X. Zhu, H. Chen, Y. Wang, L. Xia, Q. Tan, H. Li, Z. Zhong, F. Su, X.S. Zhao, Journal of Materials Chemistry A, 1 (2013) 4483-4489.

[6] Y. H. Huang, C. T. Chang, Q. Bao, J. G. Duh, Y. L. Chueh, Journal of Materials Chemistry A, 3 (2015) 16998-17007.

[7] J. H. Lee, W. J. Kim, J. Y. Kim, S. H. Lim, S. M. Lee, Journal of Power Sources, 176 (2008) 353-358.

[8] S.Y. Kim, J. Lee, B. H. Kim, Y. J. Kim, K.S. Yang, M. S. Park, Acs Applied Materials & Interfaces, 8 (2016) 12109-12117.

[9] B. Fuchsbichler, C. Stangl, H. Kren, F. Uhlig, S. Koller, Journal of Power Sources, 196 (2011) 2889-2892.

[10] M. Ko, S. Chae, J. Ma, N. Kim, H. W. Lee, Y. Cui, J. Cho, Nature Energy, 1 (2016).

[11] S. H. Yook, S. H. Kim, C. H. Park, D. W. Kim, Rsc Advances, 6 (2016) 83126-83134.

[12] X. Dong, C. Lu, L. Wang, P. Zhou, D. Li, L. Wang, G. Wu, Y. Li, Rsc Advances, 6 (2016) 12737-12743.

[13] H. Wang, J. Xie, S. Zhang, G. Cao, X. Zhao, Rsc Advances, 6 (2016) 69882-69888.

[14] B. Lu, B. Ma, R. Yu, Q. Lu, S. Cai, M. Chen, Z. Wu, K. Xiang, X. Wang, Chemistryselect, 2 (2017) 3479-3489.

[15] M. Q. Li, M. Z. Qu, X. Y. He, Z. L. Yu, Electrochimica Acta, 54 (2009) 4506-4513.

[16] M. Su, Z. Wang, H. Guo, X. Li, S. Huang, W. Xiao, L. Gan, Electrochimica Acta, 116 (2014) 230-236.

[17] Z. Wang, Z. Mao, L. Lai, M. Okubo, Y. Song, Y. Zhou, X. Liu, W. Huang, Chemical Engineering Journal, 313 (2017) 187-196.

[18] J. Zhang, Y. Liang, Q. Zhou, Y. Peng, H. Yang, Journal of Power Sources, 290 (2015) 71-79.

[19] H. Zhao, A. Du, M. Ling, V. Battaglia, G. Liu, Electrochimica Acta, 209 (2016) 159-162.
[20] J. Wu, Z. Zhu, H. Zhang, H. Fu, H. Li, A. Wang, H. Zhang, Z. Hu, Journal of Alloys and Compounds, 596 (2014) 86-91.