Supporting Information

Large size nitrogen-doped graphene-coated graphite for high performance lithium-ion battery anode

Xiaoxu Liu, Erjia Liu, Dongliang Chao, Liang Chen , Shikun Liu, Jing Wang, YaoLi, Jiupeng Zhao, Yong-Mook Kang , Zexiang Shen

Electrochemical measurements

The electrochemical tests were carried out using a coin-type cell (CR 2032). For LGAs and pure N-doped graphene electrodes, the working electrodes were prepared by mixing 90wt% active material, 5wt% acetylene black, and 5 wt% polyvinylidene fluoride binder dissolved in N-methyl-2-pyrrolidinone. After coating the above slurries on Cu foils, the electrodes were dried at 80°C in a vacuum for 2 h to remove the solvent before pressing. Then, the electrodes were cut into disks (diameter of 12 mm) and dried at 100°C for 24 h in a vacuum. The mass of the LGA composite electrode was ~3 mg. The battery cells were assembled in an argon-filled glove box with the metallic lithium foil as the counter electrode, 1 M $LiPF_6$ in ethylene carbonate (EC)–dimethyl carbonate (DME) (1:1 in volume) as the electrolyte, and a polypropylene (PP) film (Celgard 2400) as the separator. Full cells were fabricated by using a $LiCoO_2$ cathode and a LGA anode. The cyclic voltammetry (CV) measurements of pure graphite and LGA anode were carried out using a Solartron 1287 electrochemical workstation at a scanning rate of 0.1 mV s^{-1}. For electrochemical impedance spectroscopy (EIS), the amplitude of the sine perturbation signal was 5 mV, and the frequency was scanned from the highest (100 kHz) to the lowest (10 mHz). Galvanostatic charge–discharge cycles were tested on a LAND CT2001A electrochemical workstation at a current density of 500 mA g^{-1} between 0.05 and 1.5 V vs Li^+/Li at room temperature.

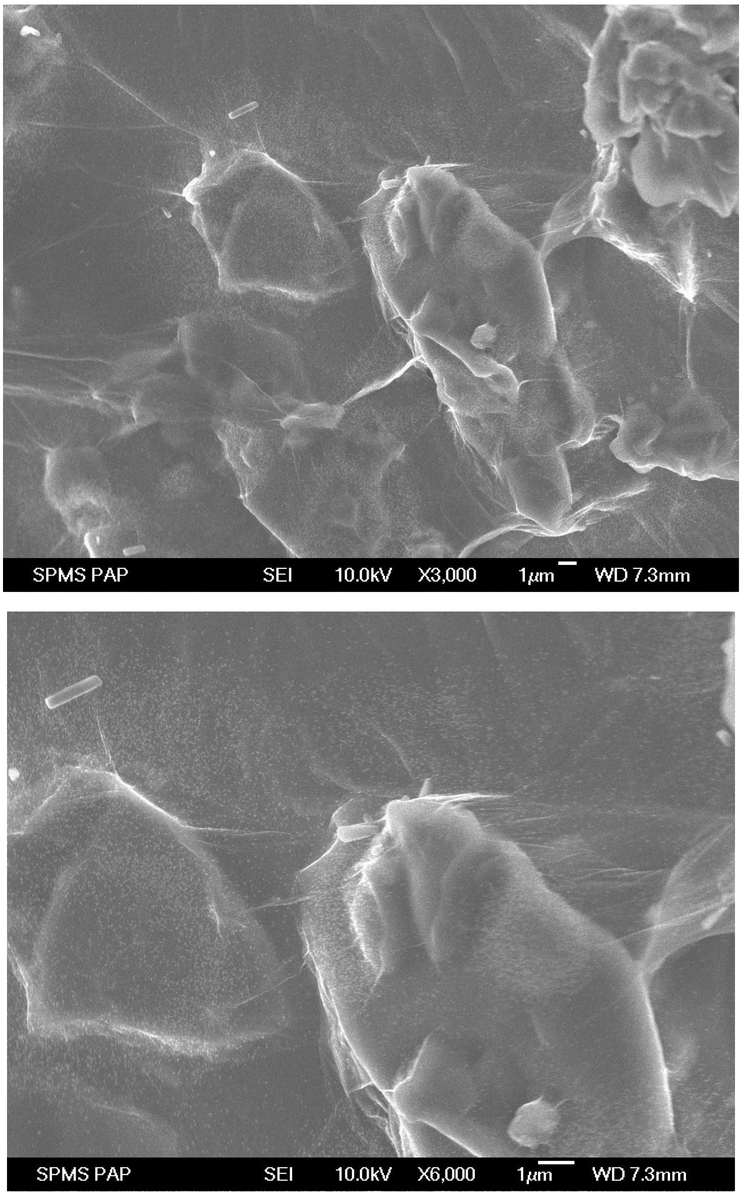

Figure S1.SEM images of LGA composite with 5wt% LNG measured with different magnifications.

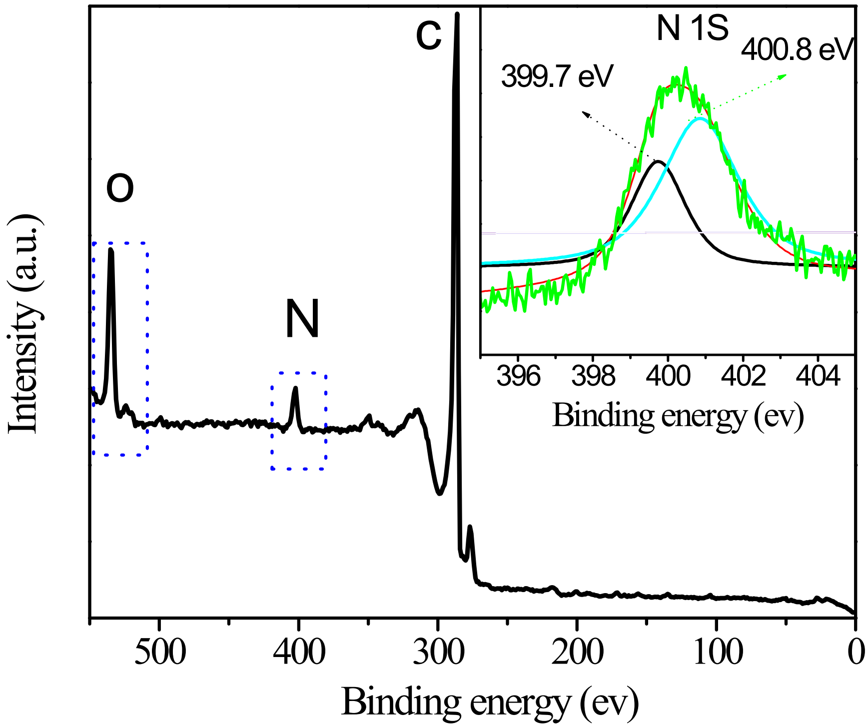

Figure S2. XPS spectra of a LGA anode with 1 wt% LNG

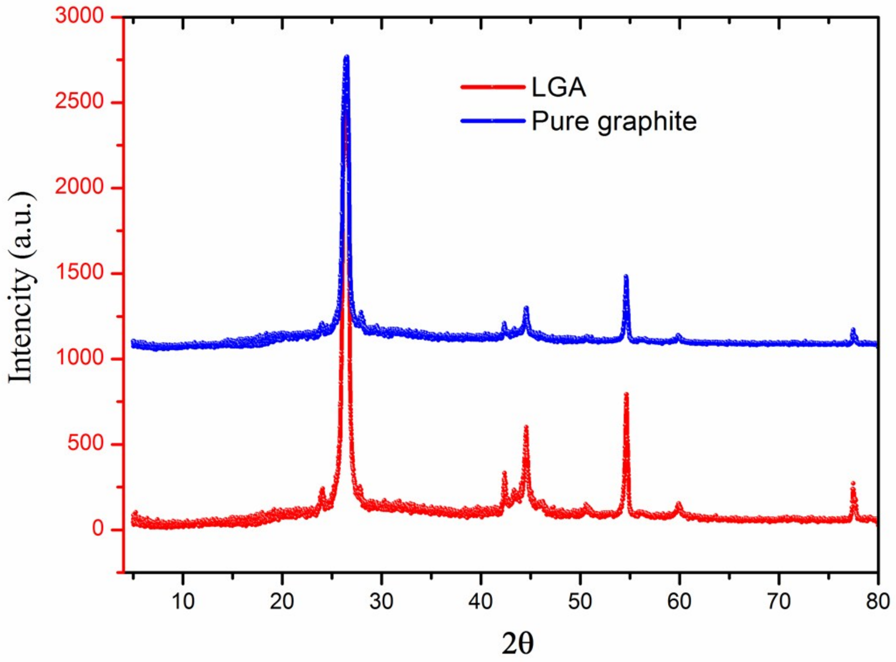

Figure S3. XRD patterns of the LGA and pure graphite

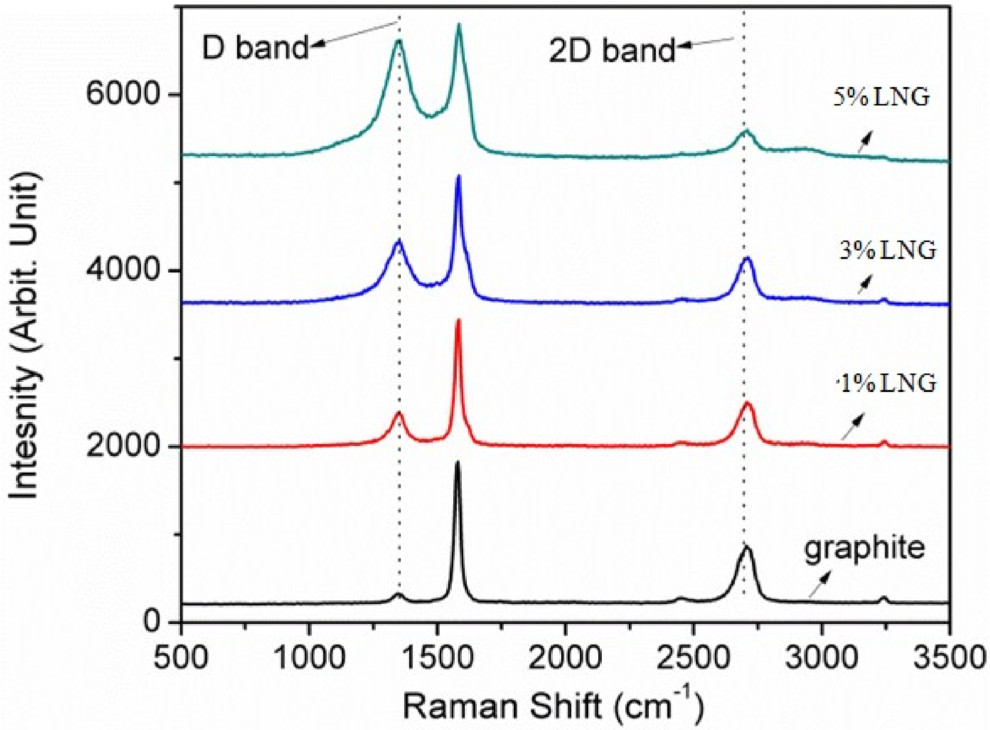

Figure S4. Raman spectra of LGA s with 0, 1, 3, and 5 wt% LNG contents.

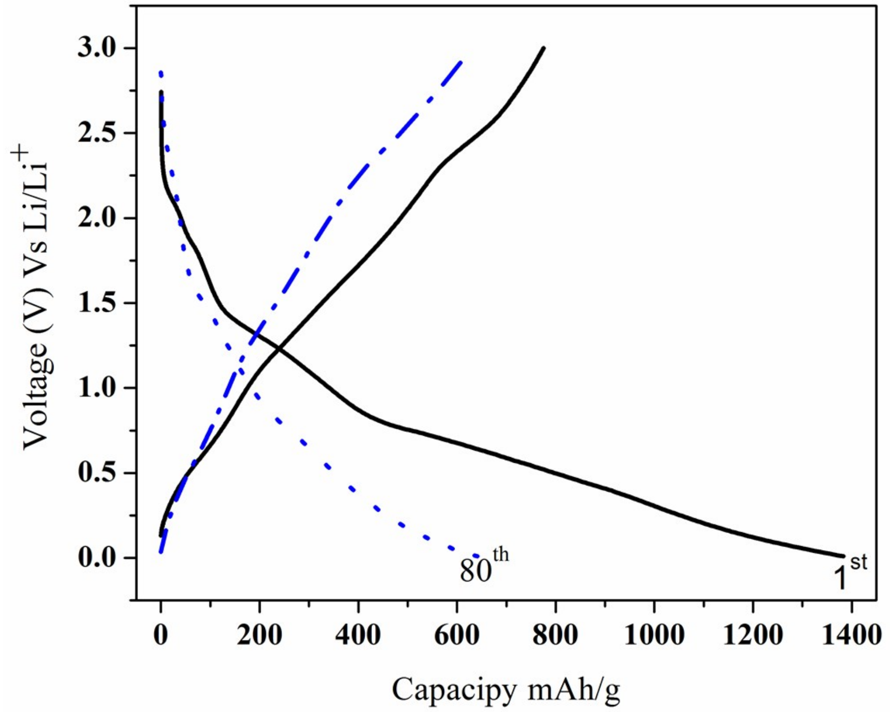

Figure S5. The discharge/charge curves of pure LNG

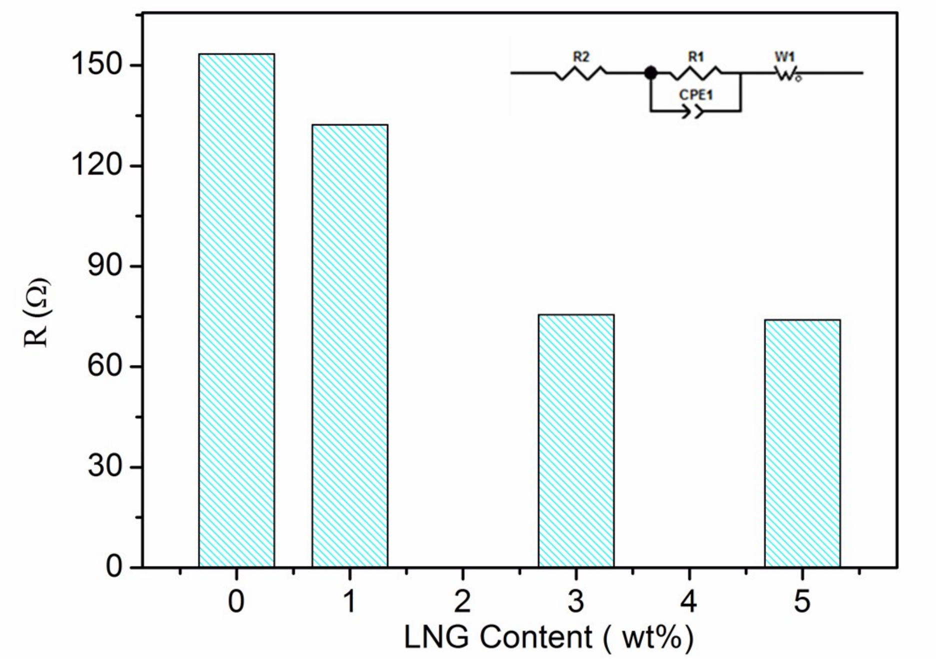

Figure S6. The contact impedances of LGA with 0, 1, 3, and 5% LNG

The conductivity of the LNG and a conventional reduced graphpene sheets by annealing in H_2/Ar at high temperatures were tested by 4-point probe measurements. the LNG is more conductive (433 S cm^{-1}) than conventional reduced graphpene (356 S cm^{-1}).

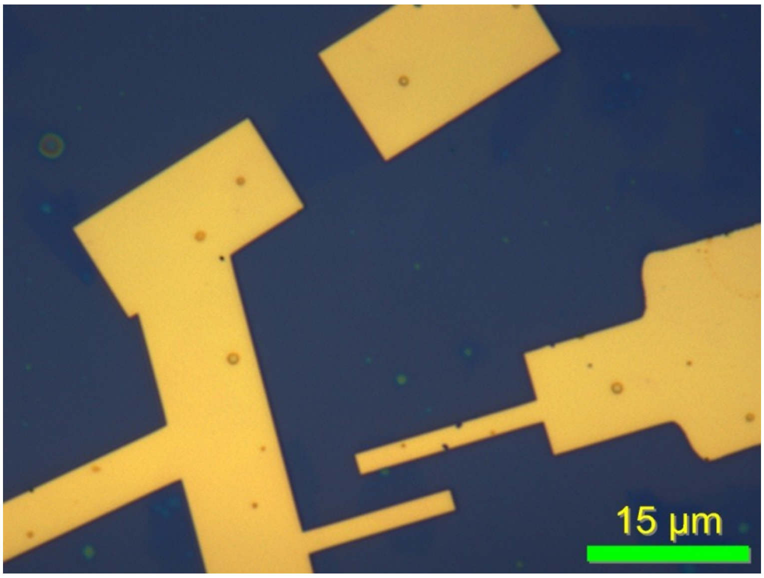

Figure S7. SEM image of LNG on the Si substrate for 4-point measurements

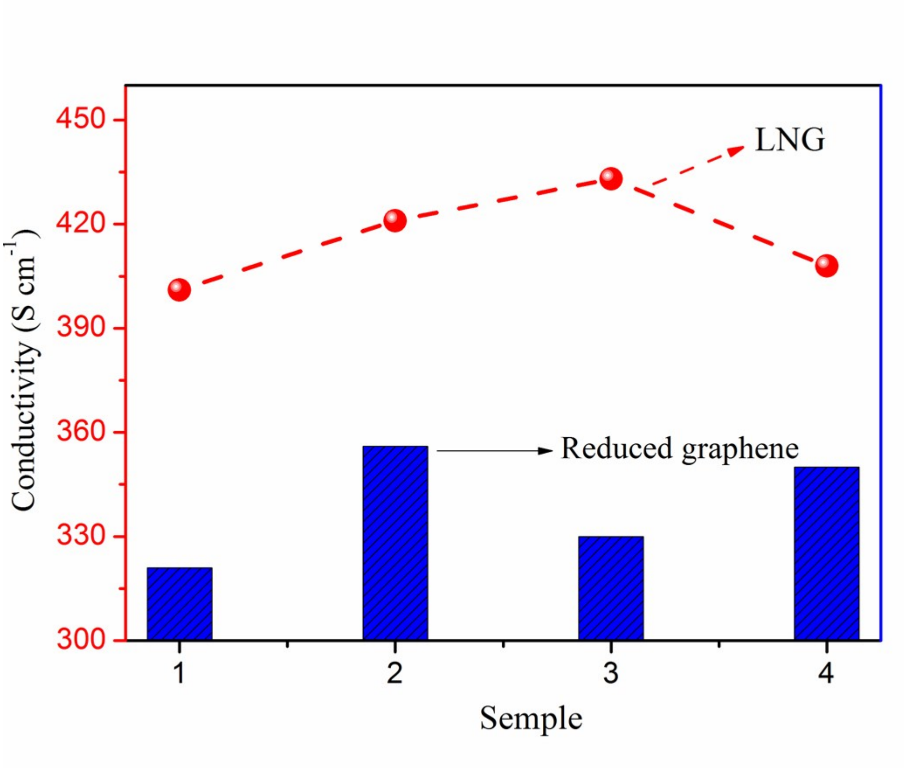

Figure S8. Plot of the mean conductivities for LNG and reduced graphene sheets

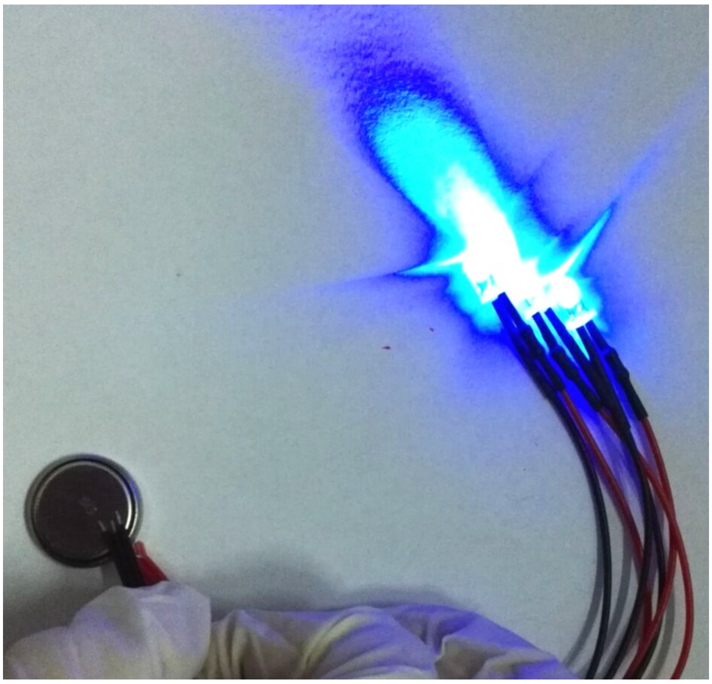

Figure S9. Digital photograph of LED powered by LGA/$LiCoO_2$ full cell

First, the size of the graphite particles used is ~ 10–100 μm. For the ease of understanding, the graphite particles are assumed to be cubical with interparticle spacing of ~ 0.33 nm (Fig.S11), which means that a single cubical graphite particle contains about 30,000 layers of square graphene sheets with side length of about 10 µm. Theoretically, to fully wrap a cubical graphite particles around 30 graphene sheets would be sufficient. Thus, it is expected that about 0.1 wt% of graphene can fully wrap the graphite particles and greatly improve the LGA performance. Conversely, excessive graphene can cause over-stacking and agglomeration of graphene, adversely affecting the composite properties. The above explanation is consistent with the experimental results obtained in this work, i.e., the capacity and initial Coulombic efficiency of the LGA anodes containing 5 wt% of graphene are lower than those of the LGA s containing 1 and 3 wt% of graphene. However, due to different size distributions of the graphite particles and the graphene platelets, the graphite-to-graphene ratio needs to be optimized through specific experiments in different systems, for better performance of the LGA composites.

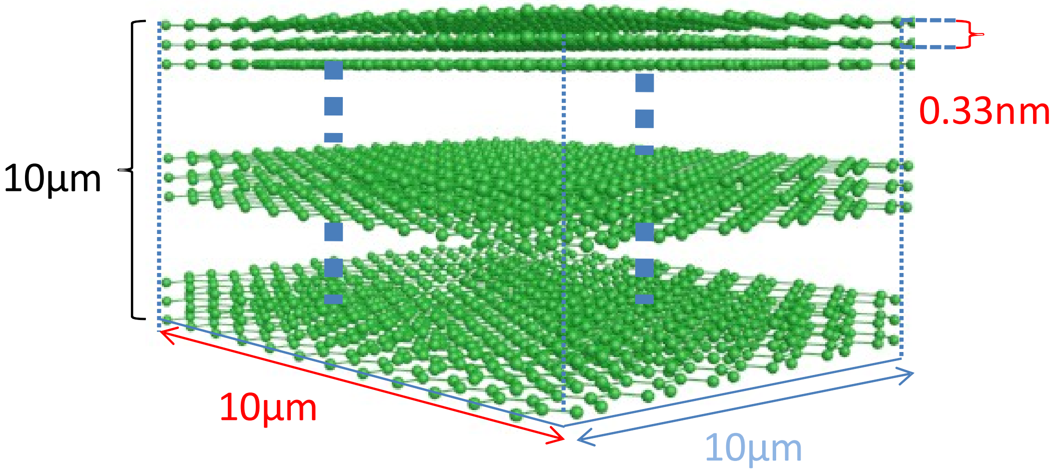

Figure S10. A model of cube-shaped graphite particles

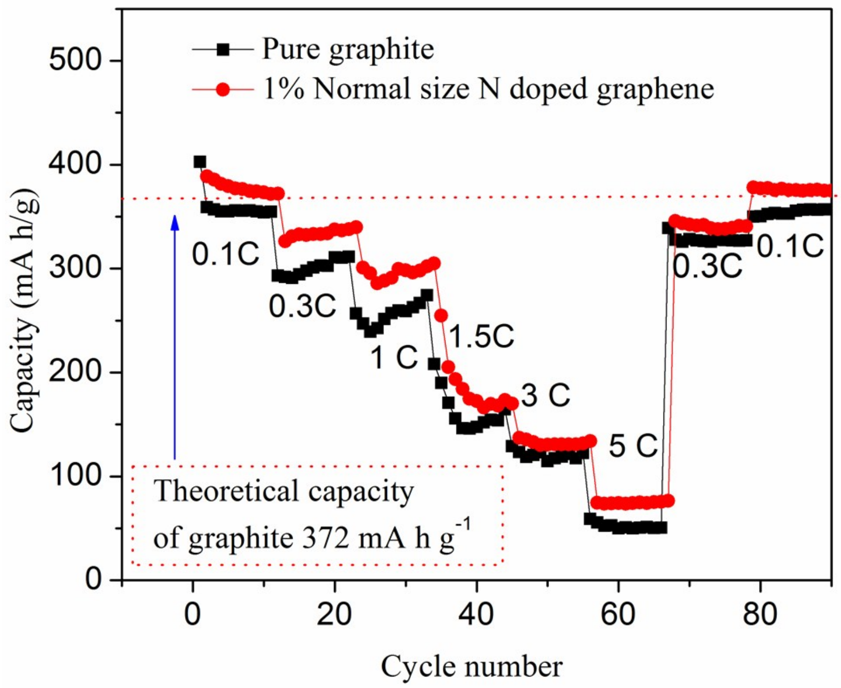

Figure S11. The rate capability of pure graphite and composite anode with 1 wt % normal size graphene,

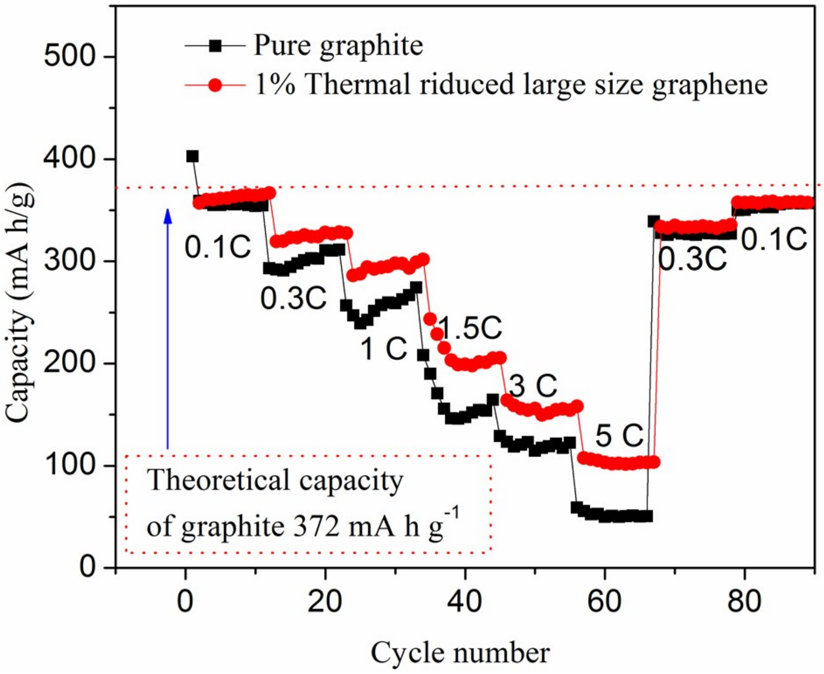

Figure S12. The rate capability of pure graphite and composite anode with 1 wt % thermal reduced large size graphene,

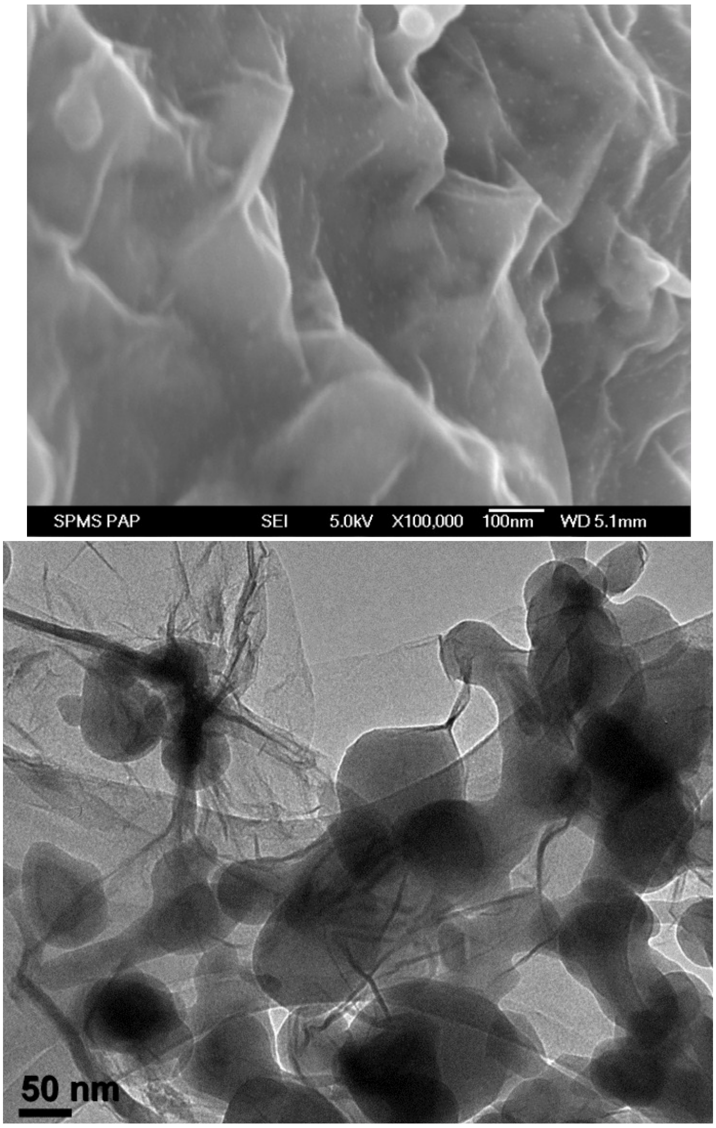

Figure S13. SEM and TEM images of LNG coated Si composite

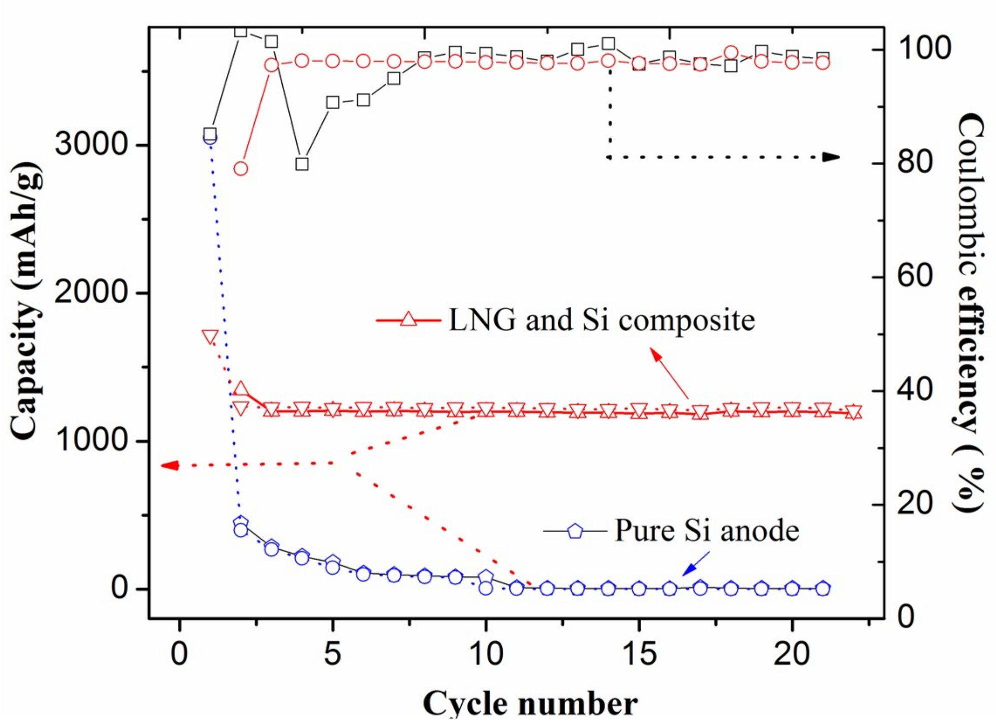

Figure S14. Cycle life test for the LIB cells on LNG coated Si anode

Table S1 A survey of electrochemical properties of surface modified graphite anode.

trode ription	*Capacity (mA hg^{-1})*	*Cycling stability*	*Initial Coulombic efficiency*	*Ref.*
LGA	390 @ 0.1 C	98% after 2000 cycles@ 1C	～89%	This work
Alkali carbonate-coated graphite electrode	*350 @ 1 C*	*50 cycles@ 1 C*	*～90%*	*1*
Reduced graphene	225 @ 0.13 C	～95% after 100 cycles @ 0.13 C	44%	2
carbon coating on natural graphite	348@ 0.1C	----	93%	3
Cu coating on graphite electrode	280@ 0.25C	----	～80%	4
Cu_2O coating on graphite	211@ 0.1mA cm^{-2}	----	～68%	5
S_b-coated mesophase graphite powder	325@ 0.1mA cm^{-2}	101.2% after 20 cycles@0.325mA cm^{-2}	～89%	6
coating with a pyrolytic carbon shell	～345@ 0.1C	90% after 40 cycles	～88%	7
Li_2CO_3-coating on the performance of natural graphite	～260@ 0.1mA cm^{-2}	80% after160 cycles	---	8

coating of the pyrolytic carbon	**～350@ 0.1mA cm^{-2}**	**95% after200 cycles @ 0.5 C**	**～88%**	9
$AlPO_4$ Coating graphite	**～360@ 0.2C**	**78% after 50 cycles**	**～93%**	10

Reference

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