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# Supporting information

# Scalable synthesis of N-doped Si nanoparticles for high-performance Li-ion batteries

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**Experimental section**

All the reagents used here are of analytical grade without further purification. Mg powder and commercial Si powder were purchased from Sinopharm Chemical Reagent Co. Ltd.

***Structural Characterization:*** The structure and morphology of the samples were characterized by X-ray diffractometer (Philips X' Pert Super diffract meter with Cu Kα radiation (λ=1.54178 Å)), Raman spectrometer (Lab-RAM HR UV/VIS/NIR), X-rayphotoelectron spectroscopy (XPS) (ESCA-Lab MKII X-ray photoelectron spectrometer), scanning electron

microscopy (SEM, JEOL-JSM-6700F), transmission electron microsocopy (TEM, Hitachi H7650 and HRTEM, JEOL 2010), and atomic resolution analytical microscope (JEM-ARM200F).

***Electrochemical Measurement:*** The electrochemical properties of the commercial Si (200 mesh) and the as-prepared N-doped Si nanoparticles were evaluated through coin-type half cells (2016 R-type) which were assembled under an argon-filled glove box ($H_2O$, $O_2$< 1 ppm).Metallic Li sheet was used as counter and reference electrode. 1 M $LiPF_6$ in a mixture of ethylene carbonate/dimethylcarbonate (EC/DMC; 1:1 by volume) was served as the electrolyte (Zhuhai Smoothway Electronic Materials Co., Ltd (China)). For preparing working electrode, the slurry mixed with as-prepared active material, carbon black (super P) and sodium carboxymethyl cellulose (CMC) binder in a weight ratio of 6:2:2 in water solvent was pasted onto a Cu foil and then dried in a vacuum oven at 80 $^oC$ for 10 h. The active material density of each electrode was determined to be about 0.8 mg $cm^{-2}$. Galvanostatic measurements were conducted using a LAND-CT2001A instrument with a fixed voltage range of 0.015−1.5 V (vs. $Li/Li^+$) at room temperature. Electrochemical impedance spectroscopy (EIS) was performed on electrochemical workstation(CHI660D), with an alternating current (AC) voltage of 5 mV in the frequency range from100 kHz to 0.1 Hz.

**Table S1 Three repeated enlarged experiments of N-doped Si nanoparticles and corresponding conversion yields.**

| | Commercial Bulk Si | Intermediate $Mg_2Si$ | N-doped Si nanoparticles | Yield |
|---|---|---|---|---|
| 1 | 25.000g | 67.278g | 23.807g | 95.2% |
| 2 | 25.000g | 67.615g | 24.578g | 98.3% |
| 3 | 25.000g | 67.321g | 24.329g | 97.3% |

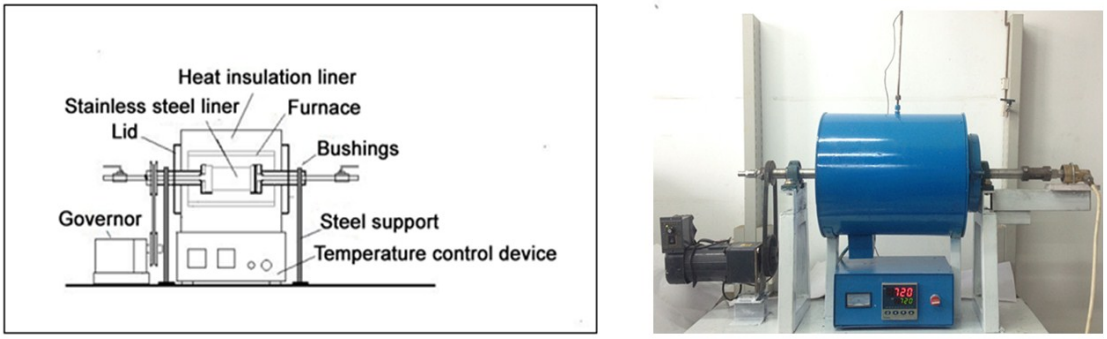


**Fig. S1. The scheme and photo of the self-designed reaction vessel.**

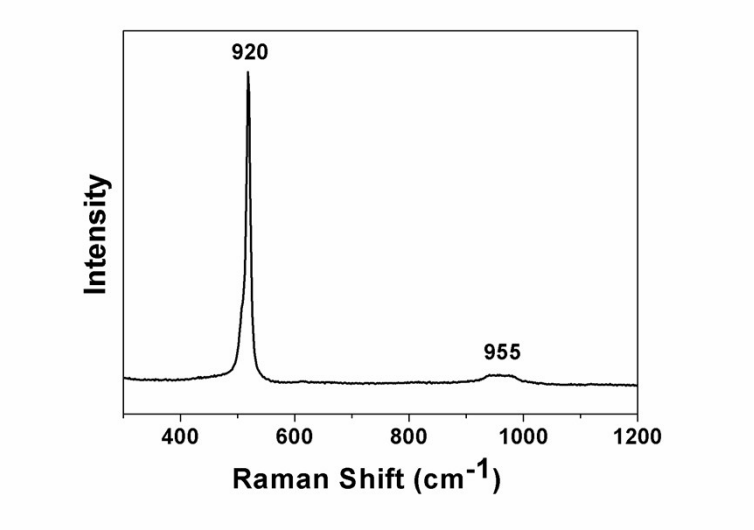


**Fig. S2. Raman spectrum of the commercial Si.**

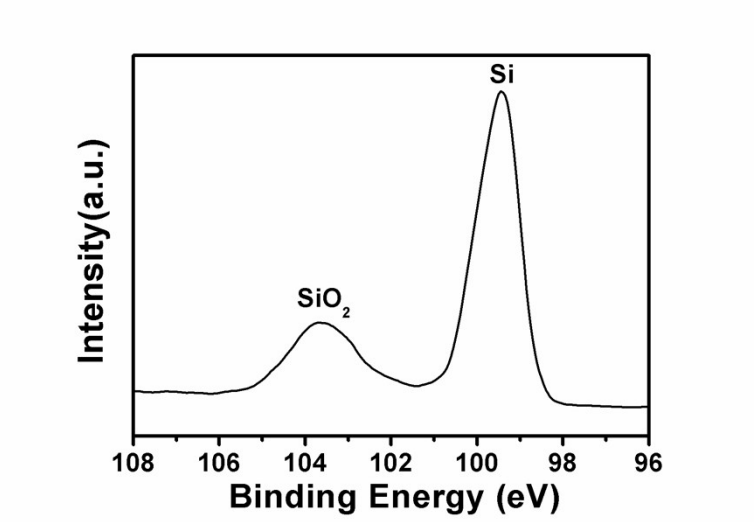


**Fig. S3. XPS spectrum of the commercial Si.**

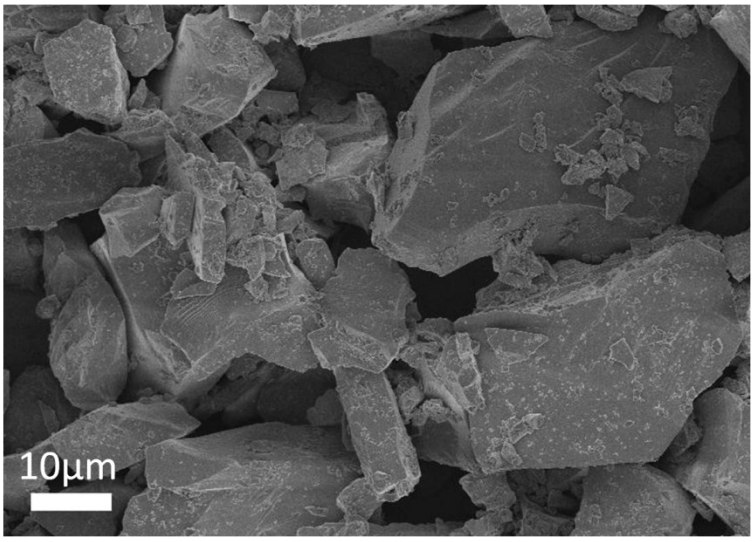


**Fig. S4. SEM of the commercial Si.**

The origin Si (200 mesh) is commercial available, and the particles size is about few micrometers (shown in Fig. S4).

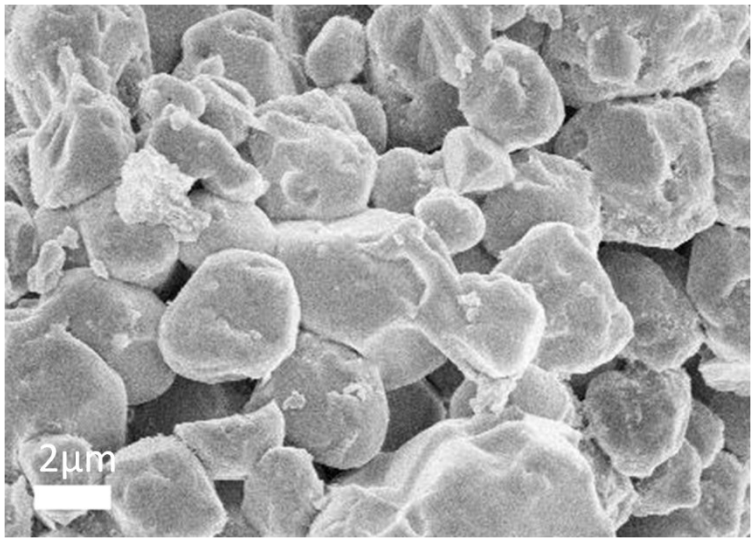


**Fig. S5. SEM of the intermediate $Mg_2Si$.**

As shown in Fig. S5, the particles size of $Mg_2Si$ becomes smaller than that of the commercial Si.

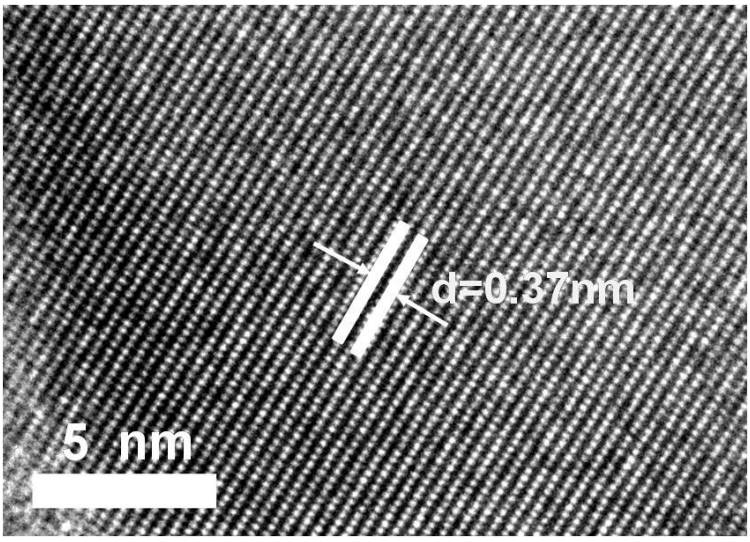


**Fig. S6. HRTEM of the intermediate $Mg_2Si$.**

The HRTEM picture shows that the interplanar distance of $Mg_2Si$ is about 0.37 nm (shown in Fig. S6), corresponding to the (111) crystal planes of the cubic phase.

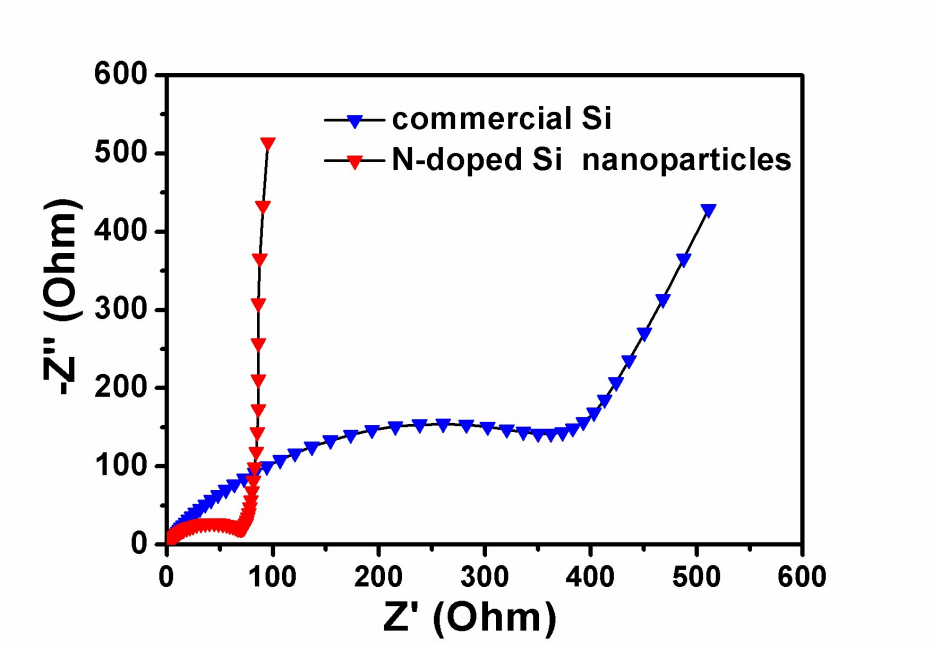


**Fig. S7.** Nyquist plots of the commercial Si and N-doped Si nanoparticles electrodes

The electrochemical impedance spectrum (EIS) is used to investigate the charge transport kinetics for the electrochemical properties of commercial Si and N-doped Si nanoparticles

electrodes. Fig. S7 shows the Nyquist plots of the AC impedance for commercial Si and N-doped Si nanoparticles electrodes. The charge transfer impedance in the electrode/electrolyte interface can be estimated by the diameter of the semicircle in the high frequency range.[1] Obviously, the commercial Si electrode shows considerably higher layer resistance than that of the N-doped Si nanoparticles electrode, which indicates that the as-prepared Si benefits the electron transfer during electrochemical reaction.

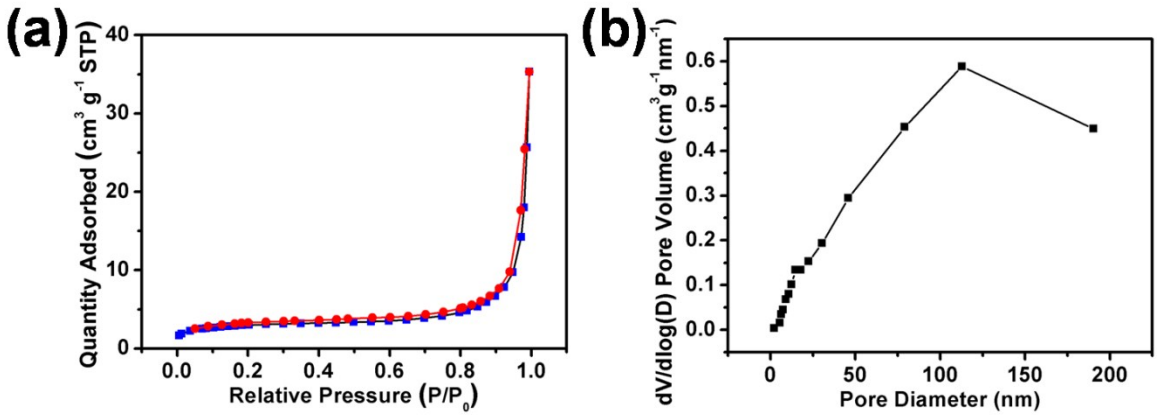


**Fig. S8. (a) Nitrogen adsorption curves, and (b) BJH pore diameter distribution of the as-prepared N-doped Si nanoparticles.**

The BET surface area of the N-doped Si nanoparticle is 10.59 $m^2$ $g^{-1}$. The $N_2$ adsorption - desorption isotherms of the N-doped Si nanoparticle is shown in Fig. S7. Refer to the SEM, TEM images of the prepared N-doped Si sample, the pore structure of N-doped Si nanoparticles could be attributed to the aggregated distribution of the nanoparticles.

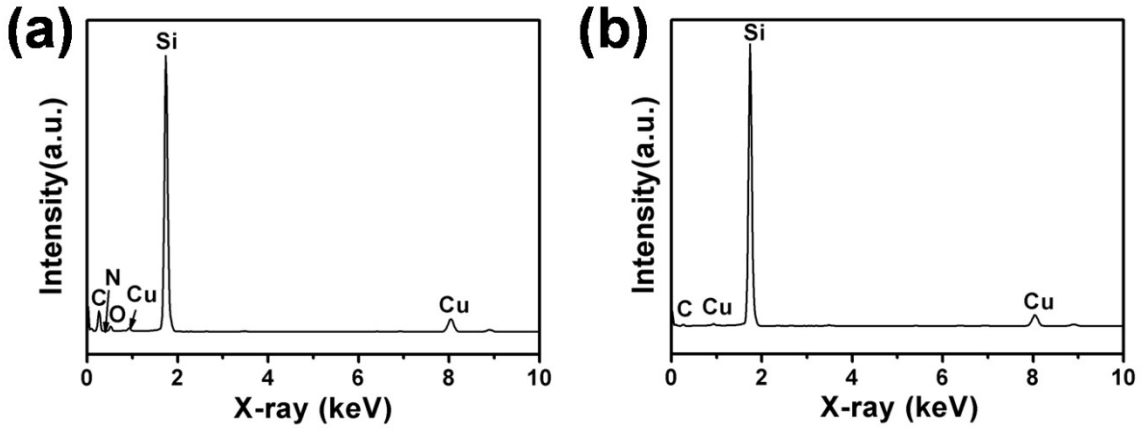


**Fig. S9. EDX spectrums of (a) N-doped Si nanoparticles, and (b) Si particles obtained under the similar conditions without the addition of Mg particles.**

As shown in Fig. S8a, the EDX spectrum of the prepared N-doped Si nanoparticles indicates that the $Mg_3N_2$ is not left in the sample after acid treatment.

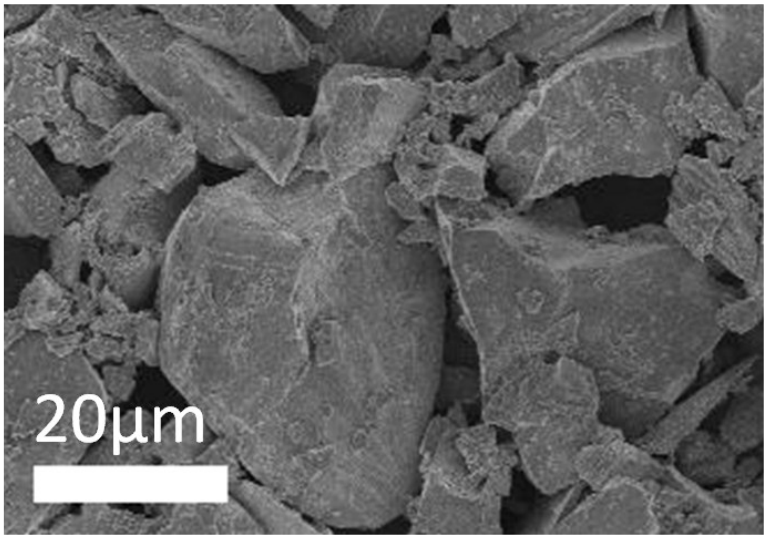


**Fig. S10. SEM of the Si particles obtained under the similar conditions without the addition of Mg particles.**

The sample has the similar size with the commercial Si.

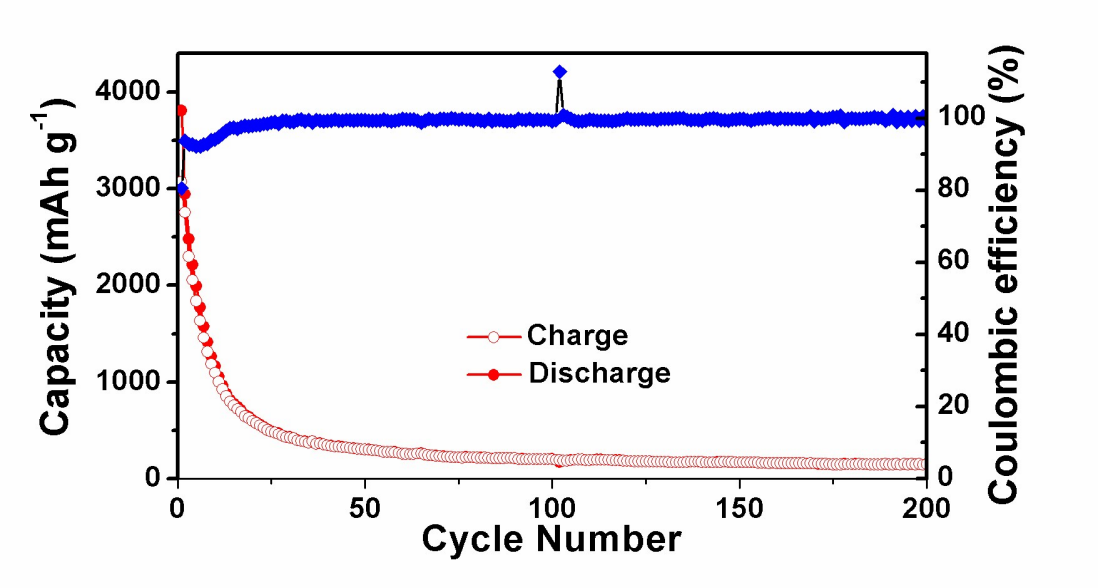


**Fig. S11. The long term cycling properties of the Si particles obtained under the similar conditions without the addition of Mg particles.**

The cycling behavior is measured at a current density of 0.36 A $g^{-1}$ (Fig. S9), the Si anode displays a specific capacity about 148 mAh $g^{-1}$ after 200 cycles.

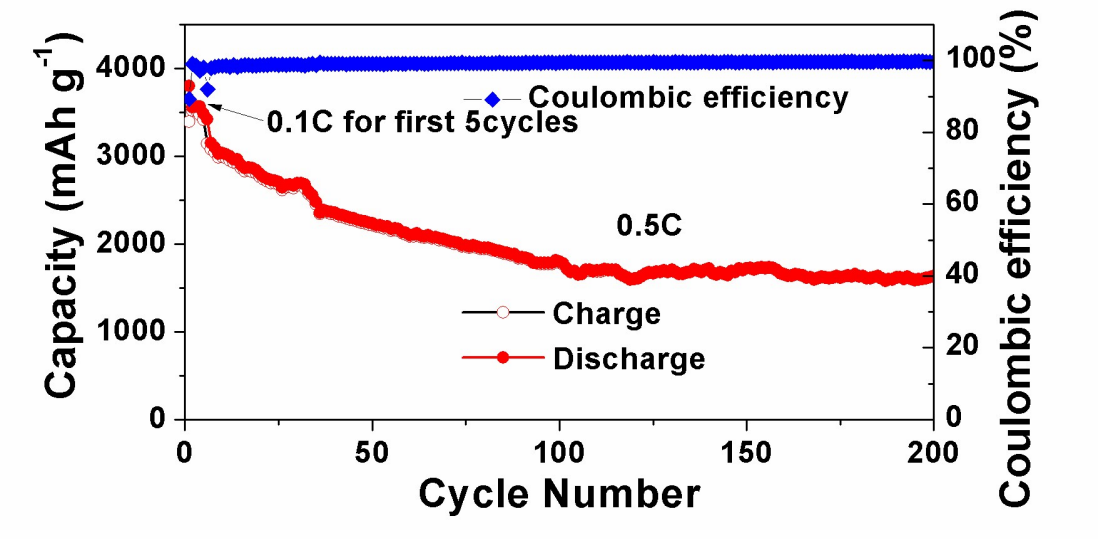


**Fig. S12. The long term cycling properties of the N-doped Si nanoparticles in enlarged experiments.**

The long term cycling behavior is measured at a current density of 1.8 A $g^{-1}$ (Fig. S8), the N-doped Si anode displays a specific capacity about 1580 mAh $g^{-1}$ after 200 cycles, which is showed excellent cyclic performance. It must be mentioned that the first five cycles of the cells are tested at a relative low current density of 0.36 A $g^{-1}$ to activate Si nanoparticles sufficiently.

**Notes and references**

1. S. L. Chou, J. Z. Wang, M. Choucair, H. K. Liu, J. A. Stride and S. X. Dou, *Electrochem. Commun.*, 2010, **12**, 303–306.