Electronic supplementary information (ESI)

Elasticity-related periodical Li storage behavior delivered by porous graphene

Guoqing Ning,* Yanming Cao, Chuanlei Qi, Xinlong Ma, Xiao Zhu

Materials	Typical	Specific	Typical cycling performance	Reference
	pore size	surface area		
	(nm)	$(m^2 g^{-1})$	500 120	
graphene and porous polyaryltriazin e derived frameworks	3.7	1683	450 Coulombic effciency for G-PPF-p-400-600 110 450 0.4A g ⁻¹ 100 350 0.4A g ⁻¹ 100 350 0.4A g ⁻¹ 100 250 G-PPF-p-400-600 60 250 G-PPF-p-600 60 251 G-PPF-p-400 60 200 G-PPF-p-400 10 200 G-PPF-p-400 30 100 G-PPF-m-400 10 20 G-PPF-o-400 10 0 100 200 300 400 500 600 700 800 Cycle number G-PPF-o-400 0 0 0 0 0 0	Figure 3b in [1]
Nitrogen- doped carbon nanotubes	Not mentione d	Not mentioned	3500 3000 2500 2500 1500 500 0 500 0 500 1000 500 1000 500 1000 1500 2500 1500 1500 0 1500 15	Figure 2c in [2]
Porous graphene	~4	1654	5 1000 1C 600 1C 1C 7 600 20 C 9 Charging 20 C 0 20 C 20 C	Figure 3c in [3]

Table S1. The gradually increased Li capacity for nanomaterials.

Materials	Typical pore size (nm)	Specific surface area $(m^2 g^{-1})$	Typical cycling performance	Reference
A conductive microporous carbon matrix, containing S in the micropores	0.5 nm	936	2000 • S/(CNT@MPC) • S/CB • Efficiency 100 (%) 50 100 • S/CB • Efficiency • S/CB • Efficiency • S/CB • CB • CD • CD	Figure 4b in [4]
Carbon shells, encapsulatin g Si nanoparticles	~100 nm	Not mentioned	3000 2500 2500 C/3 C/2 1500 C/3 C/2 1C 1.5C 2C 3C 4C 0 0 500 0 500 0 500 100 150 0 500 0 500 0 500 0 500 0 100 1.5C 2C 3C 4C 100 100 100 100 100 100 100 10	Figure 4E in [5]
Hollow carbon nanosphere	~60 nm	412	1200 (a) 1000 (a) 1000 (b) 1000 (c) 1000 (c) 1000 (c) 1000 (c) 1000 (c) 1000 (c) 1000 (c) 100 (c) 10	Figure 3a in [6]
RGO, graphene prepared via reduction of graphene oxide			1500 Charge Discharg	Figure 3b in [7]

 Table S2. The fluctuations in the cycling performance for porous carbon materials.



Figure S1. TG curve of the magnesium oxysulfate whiskers in an oxygen flow. The weight loss at ~ 314 °C corresponds to the release of crystal water in $5Mg(OH)_2 \cdot MgSO_4 \cdot 3H_2O$, and the weight loss at ~ 409 °C corresponds to the decomposition of $Mg(OH)_2$.



Figure S2. XPS survey (a) and S 2p peak (b) of PG-1.



Figure S3. Galvanostatic charge/discharge curves of the PG-1 electrode without pre-lithiation at the current density of 50 mA g^{-1} .



Figure S4. Cycling performance of a parallel sample of PG-1.



Figure S5. Illustration showing the variation of physical parameters during lithiation for a closed single nanopore (a), a semi-colosed single nanopore (b) and interconnected nanopores (c, d). In a, d and d' are the interlayer spacings before and after lithiation. In b, Φ and Φ' are the opening degrees of the entrance before and after lithiation, c_i and p_i are the Li⁺ concentration and the pressure in the nanopore, and c_0 and p_0 are the Li⁺ concentration and the pressure in the nanopore). In c and d, V and L represent volume and length, respectively.

Discussion: The closed ball shown in Figure S5a is the simplest situation for a nanopore. In that case, the interlayer placing between graphene layers will increased from *d* to *d'* due to the intercalation of Li⁺. Because the covalent bonds between C atoms are not destroyed during the lithiation, the diameter increase of the outer walls will lead to an increase of the contractility and inner pressure. The variations of Li⁺ concentration and inner pressure are shown in Figure S5b. The openning degree of the entrance (Φ) probably decreases after the lithiation because of the formation of SEI films and the expansion of the nanopore. The pressure in the lithiated nanopore (p'_i) should be higher than that before lithiation (p_i), because of the increased tensile force of the pore wall and the decreased openning degree. Because of the

good adsorpbality of the nanopore, the Li⁺ concentration in the nanopre (c_i) should be larger than that in the bulk eletrolyte (c_0). After lithiation, c'_i is larger than c_i , because of the Li⁺ insertion. In the PG, almost all nanopores are interconnected with each other. Therefore, the cases shown in Figure S5c and d are closer to the actual situation. Obviosly, the pore volumes of the interconnected pores will increase after lithiation, aroused by the streching of outer walls. Therefore, it is concluded that the volume and surface area of the nanopore, the Li⁺ concentration in the pore, and the the internal pressure will increase after the lithiation.



Figure S6. Rate capability of the PG-1 electrode at the current from 1 to 10 A g⁻¹.



Figure S7. Low (a), middle (b & c) and high magnification (d) TEM images of PG-2 after cycling tests.

Discussion: As shown in Figure S7a, the 3D fibrous structure of PG-2 has been well kept after cycling. Blocking and reducing of the openings of nanopores due to the deposition of SEI films and the attachment of binder, thus forming semi-closed nanopores, are observed in the material, as indicated by the arrows in Figure S7b & c. Thick SEI films are observed on the surface of the porous graphene (Figure S7d).



Figure S8. Cycling performances of PG-1 at different pressure: 2 (a), 4 (b), 6 (c), 8 (d) and 10 (e) MPa at the current density of 1 A/g. (f) The standard deviation of the coulombic efficiency (CE) at different pressures.



Figure S9. (a) AC impedance curves and (b) charge-transfer resistances (R_{ct}) of the PG-1 electrode pressed at different pressures. The inset of b is the equivalent circuit for fitting of the impedance curves. In the equivalent circuit, R_e represents the total resistance of electrolyte, electrode, and separator. R_f and CPE₁ are the resistance and capacitance (expressed by a constant phase element) of the SEI films formed on the electrode. R_{ct} and CPE₂ represent the charge-transfer resistance and the double layer capacitance, respectively, and Z_w is the Warburg impedance related to the diffusion of lithium ions into the bulk electrode.

Discussion: The charge-transfer resistance (Rct) significantly increases at the pressure of 10 MPa, indicating that the porous structure of PG-1 might be damaged.



Figure S10. Cycling performance of the PG-1 electrode with different PVDF contents: lithiation-delithitation capacities (a), CEs (b) and the standard deviations of CEs (c).

Reference

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