

Electronic Supporting Information

**Reconstructing ZnO quantum dot assembled tubular structures from nanotubes within graphene matrix *via* ongoing pulverization towards high-performance lithium storage<sup>†</sup>**

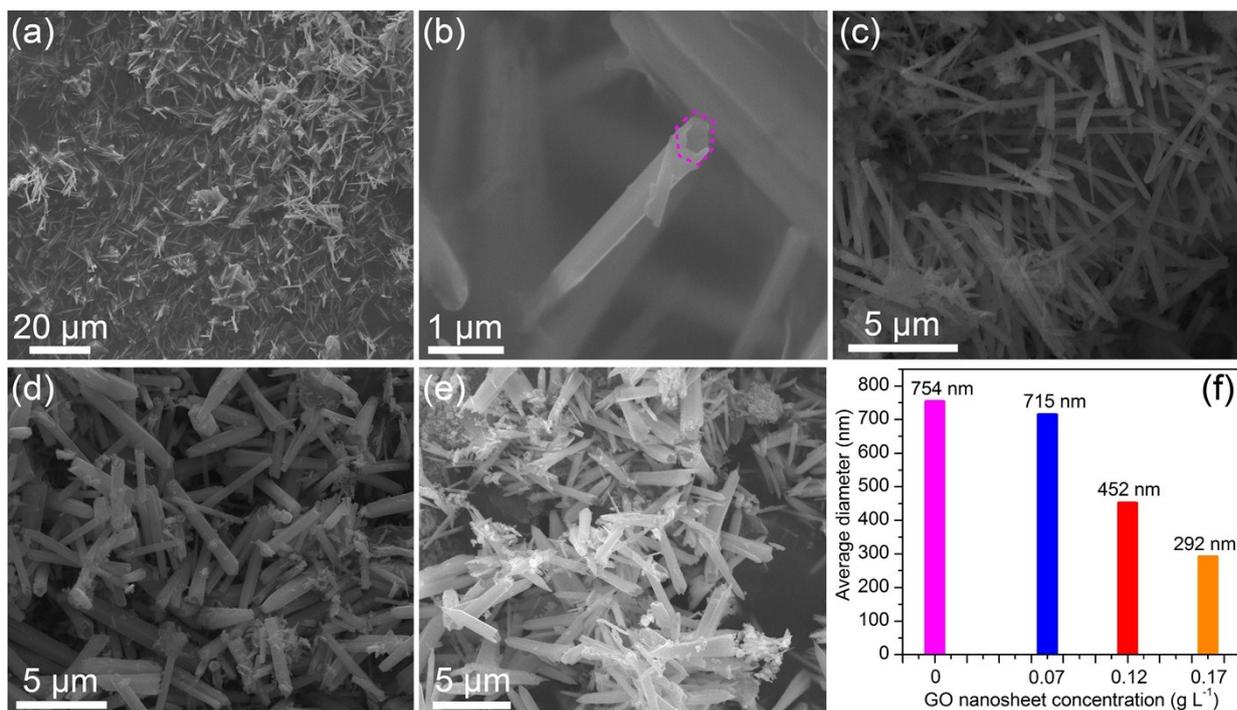
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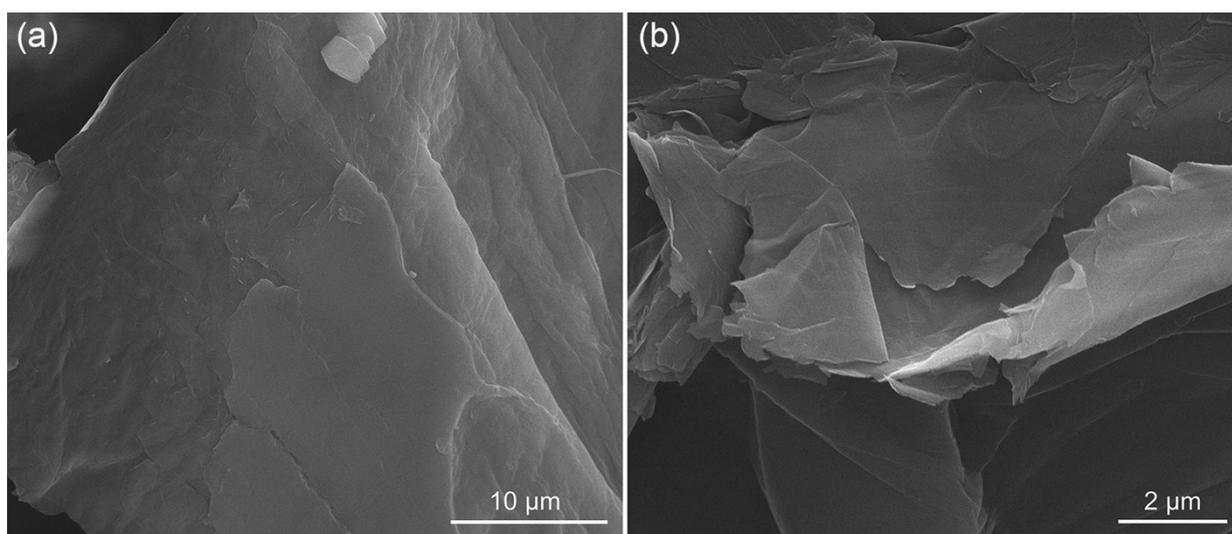
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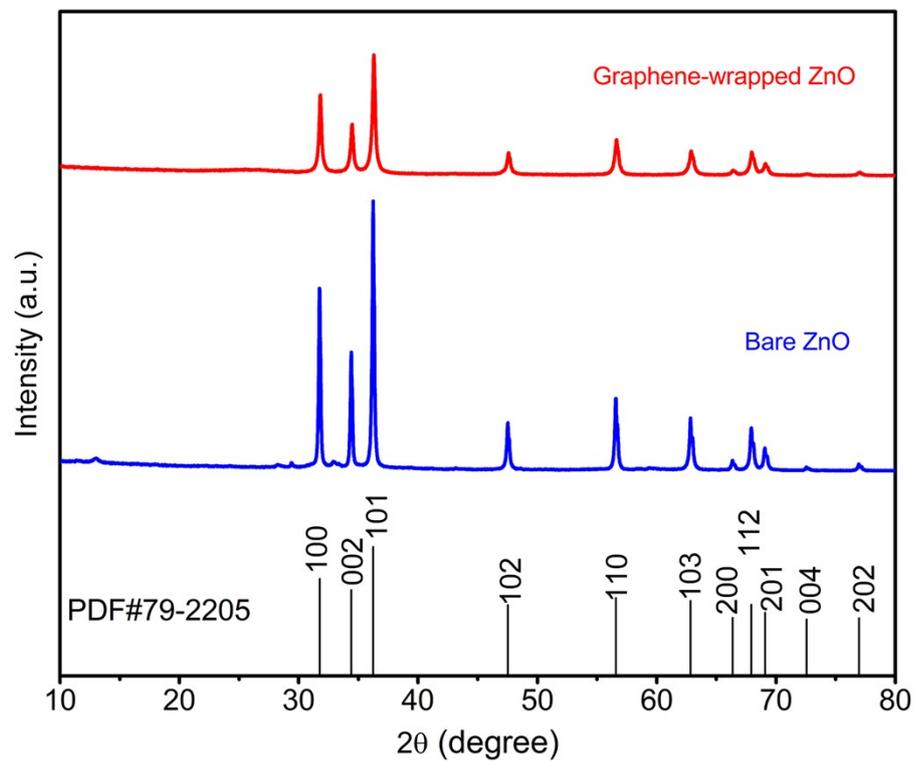
*E-mail: liuyong7@mail.sysu.edu.cn*



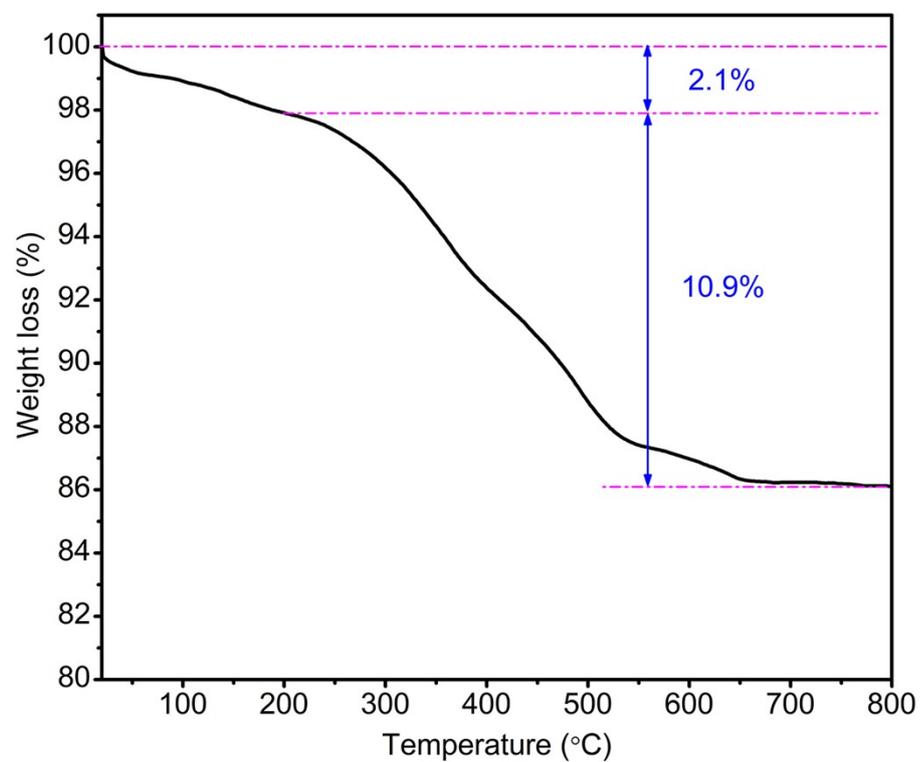
**Fig. S1** FESEM images and corresponding histogram showing the morphological evolution of ZnO nanotubes by adding different concentration of GO nanosheets in reaction solution. (a) and (b) 0.17 g L<sup>-1</sup>, (c) 0.12 g L<sup>-1</sup>, (d) 0.06 g L<sup>-1</sup>, (e) 0 g L<sup>-1</sup>, and (f) The histogram showing relations of average diameters of ZnO nanotubes summarized in above FESEM images and concentration of GO nanosheets in the reaction.



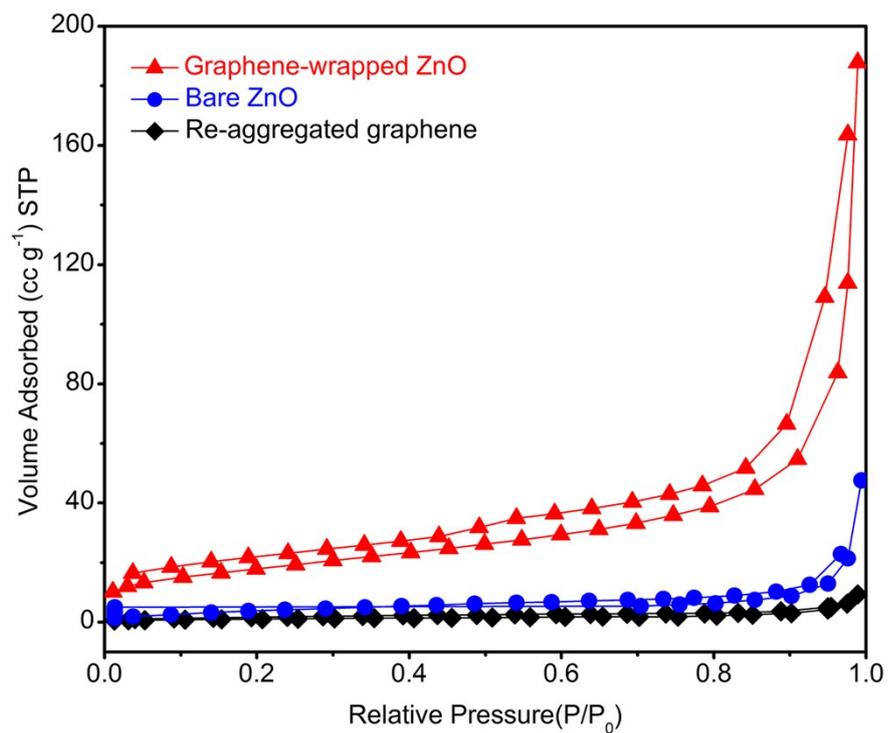
**Fig. S2** FESEM images of (a) as-synthesized graphene oxide nanosheets and (b) graphene nanosheets obtained by after reduction heat treatments at 700 °C for 2 h under 5% hydrogen in nitrogen atmosphere.



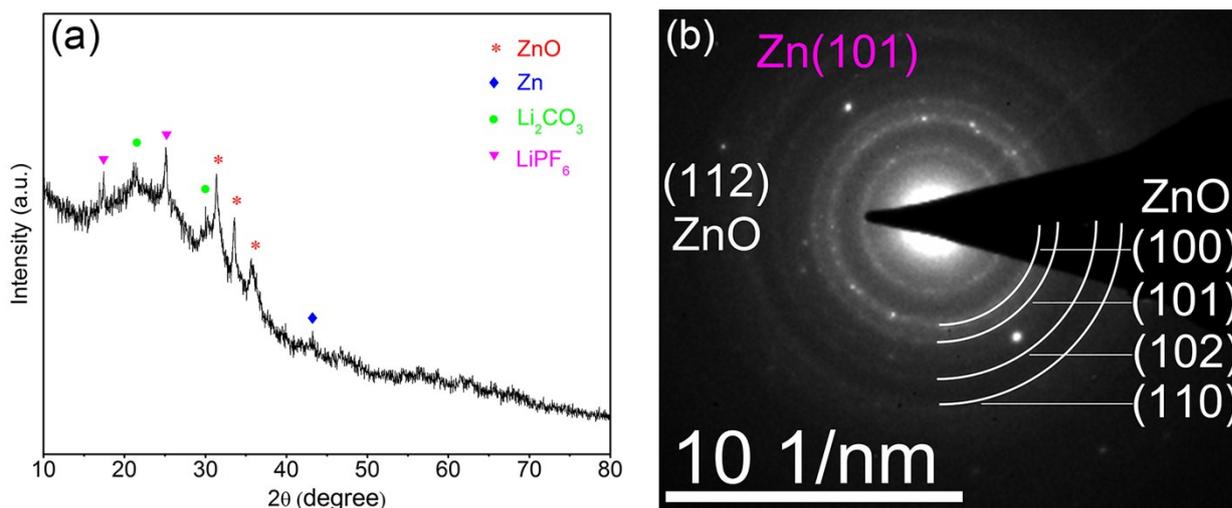
**Fig. S3** XRD patterns of post-annealed graphene-wrapped ZnO nanotubes and bare ZnO nanotubes.



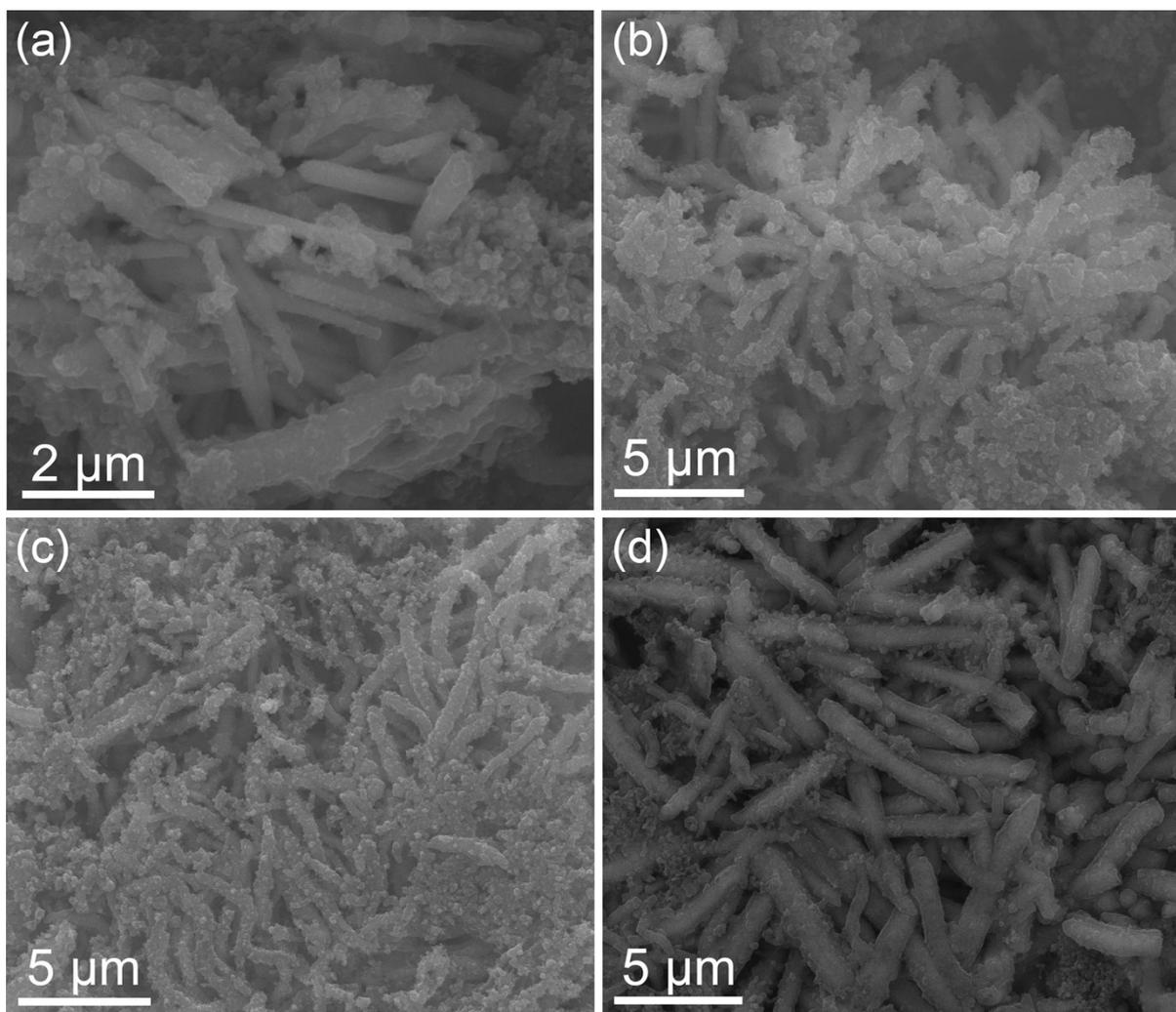
**Fig. S4** The TGA curve of the post-annealed graphene-wrapped ZnO.



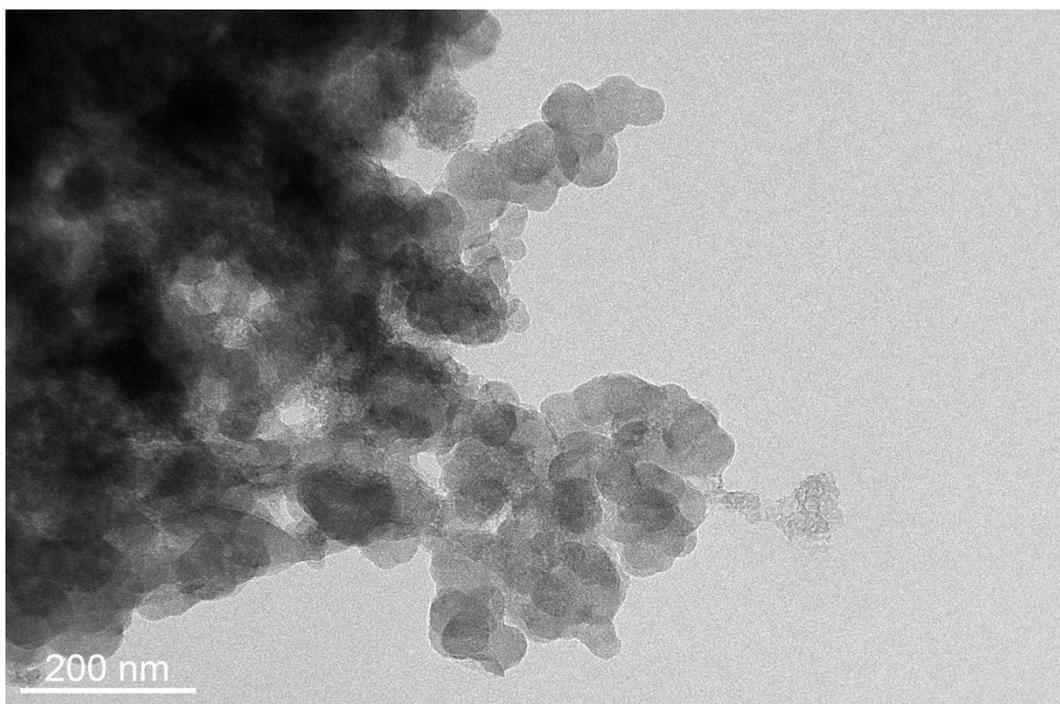
**Fig. S5** N<sub>2</sub> adsorption-desorption isotherm of the post-annealed graphene-wrapped ZnO, bare ZnO and re-aggregated graphene.



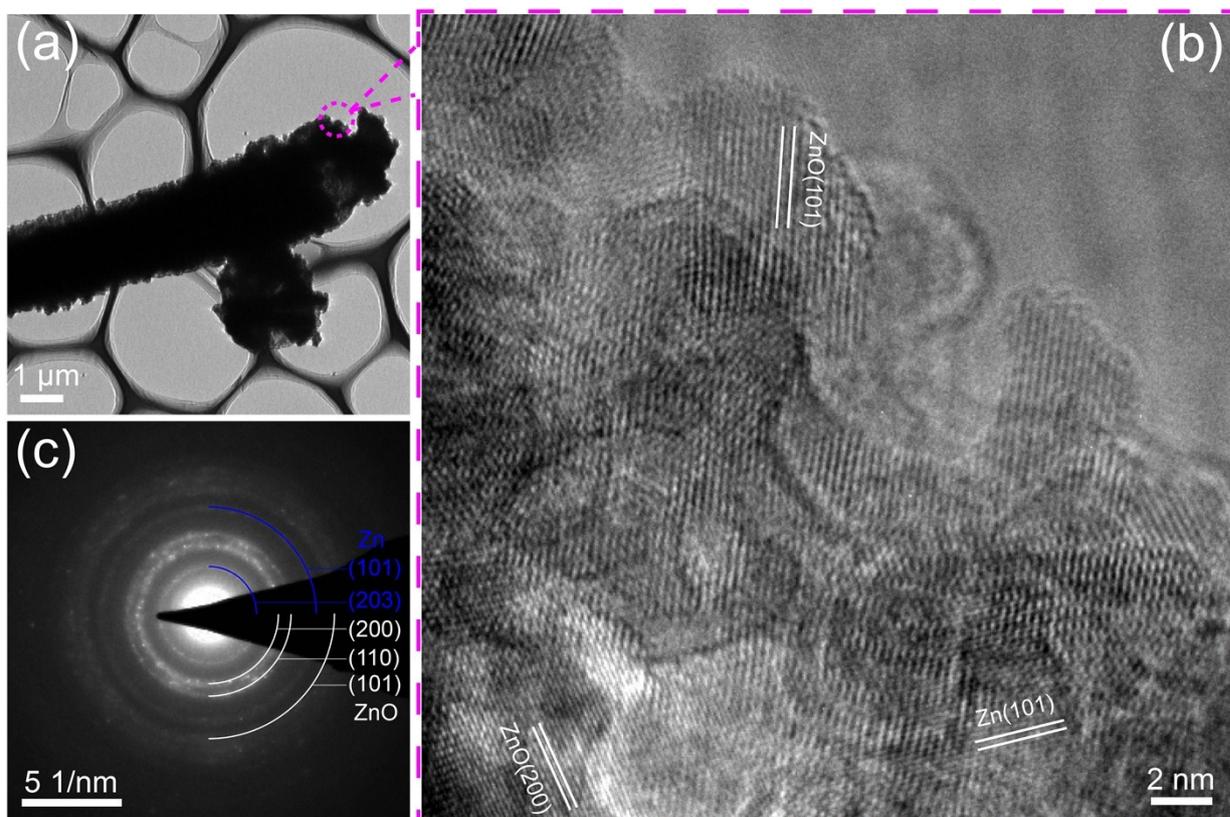
**Fig. S6** (a) XRD pattern and (b) SAED of the graphene-wrapped ZnO electrode after 1000 discharge/charge cycles at a current density of 2000 mA g<sup>-1</sup>.



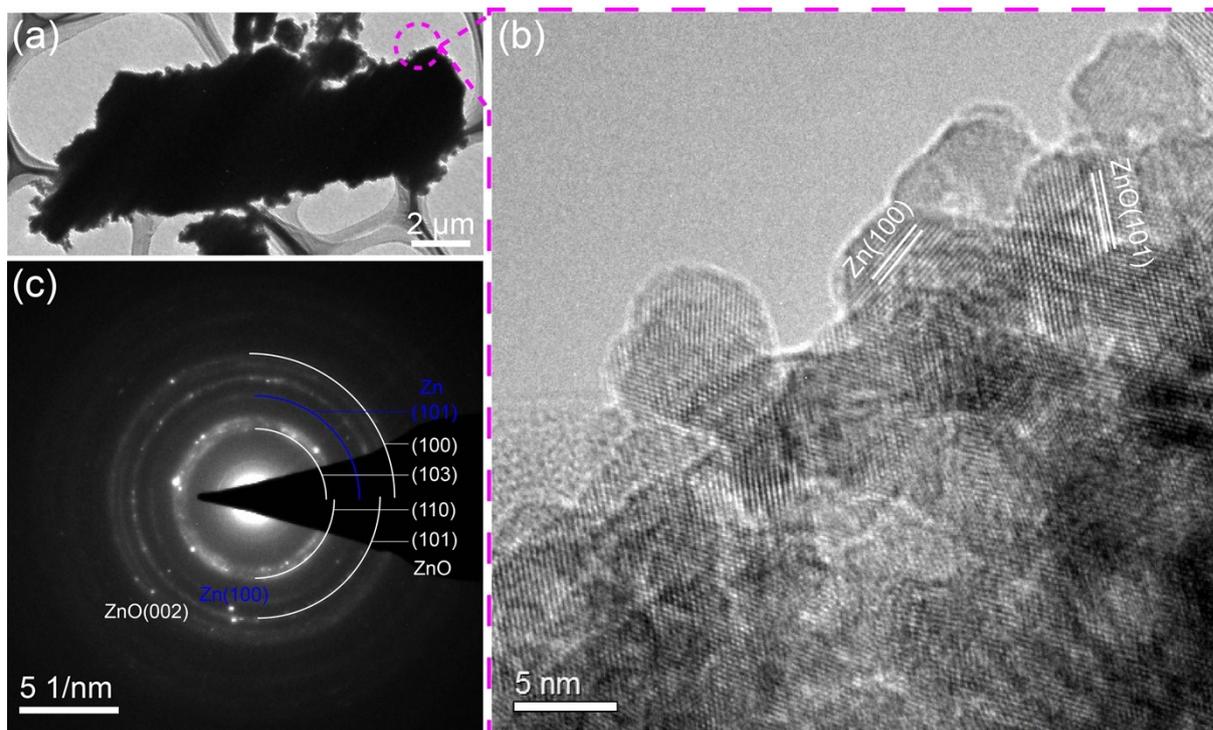
**Fig. S7** FESEM images of graphene-wrapped ZnO electrodes after (a) 2, (b) 100, (c) 400 and (d) 700 discharge/charge cycles at a current density of  $2000 \text{ mA g}^{-1}$ .



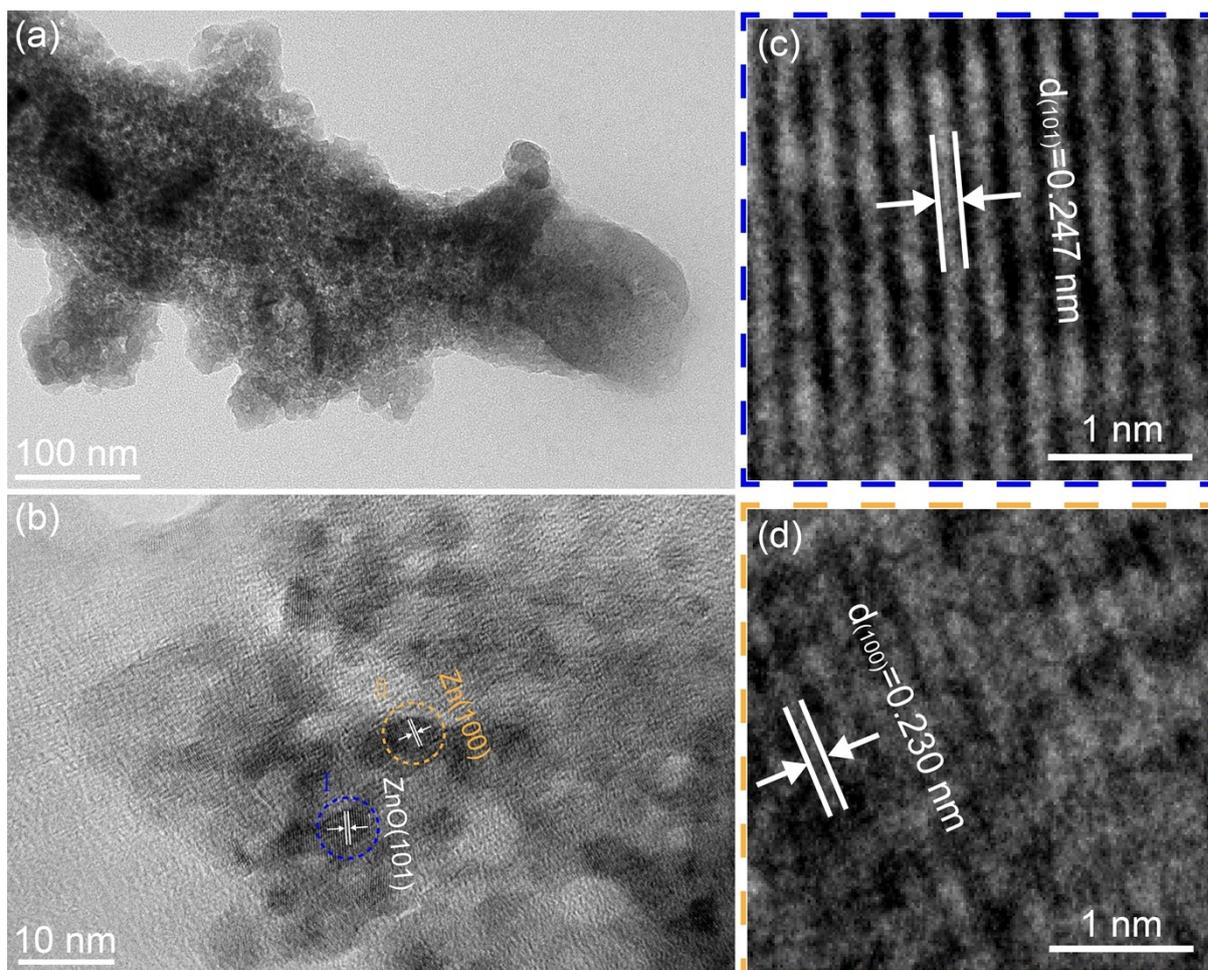
**Fig. S8** TEM image of the graphene-wrapped ZnO electrode after 100 discharge/charge cycles at a current density of  $2000 \text{ mA g}^{-1}$ .



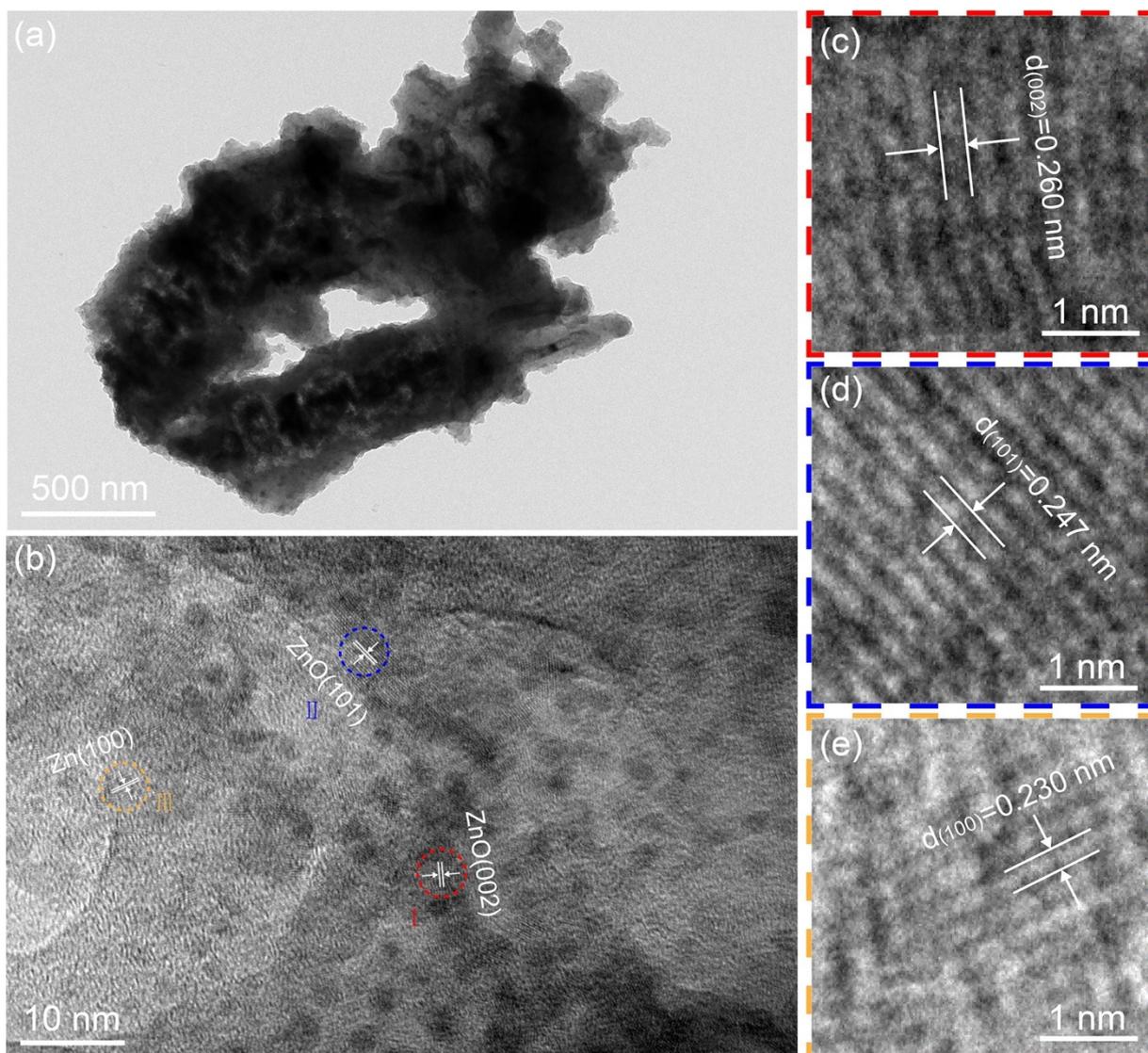
**Fig. S9** (a) The TEM image, (b) HRTEM image and (c) SAED of the graphene-wrapped ZnO electrode after 400 discharge/charge cycles at a current density of  $2000 \text{ mA g}^{-1}$ .



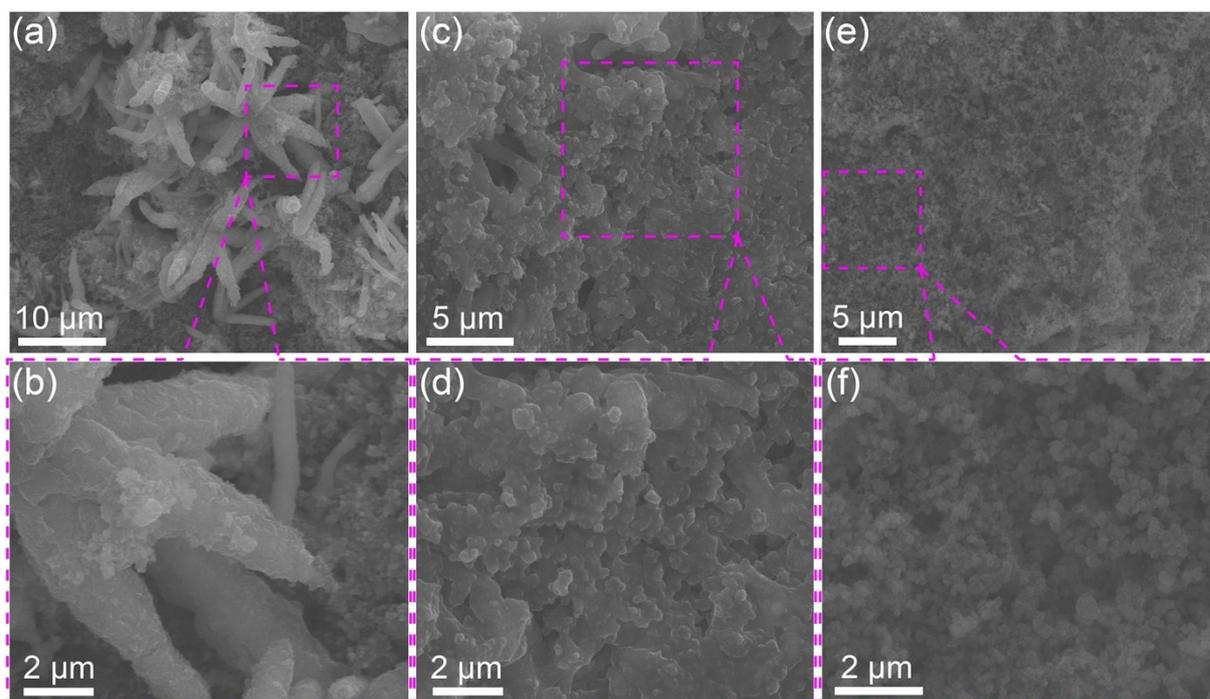
**Fig. S10** (a) The TEM image, (b) HRTEM image and (c) SAED of the graphene-wrapped ZnO electrode after 400 discharge/charge cycles at a current density of 2000 mA g<sup>-1</sup>.



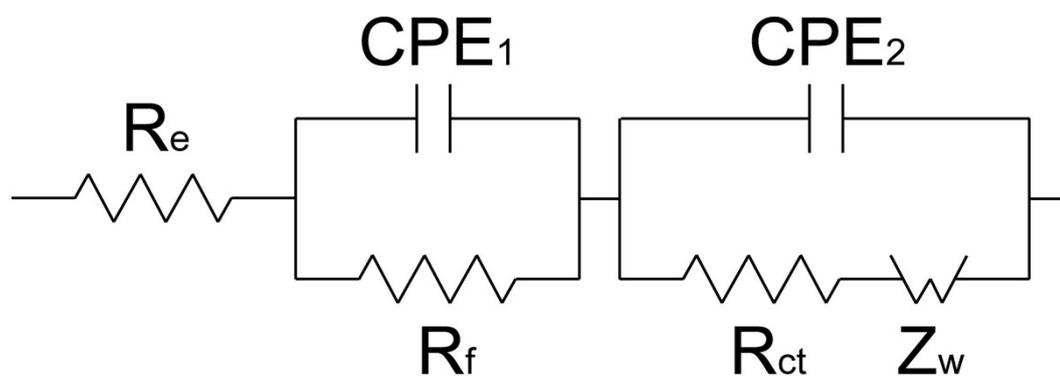
**Fig. S11** TEM images of the graphene-wrapped ZnO quantum dots assembled tubular structure after completion of 1000 discharge/charge cycles at a current density of  $2000 \text{ mA g}^{-1}$ . (a) TEM and (b) HRTEM images. (c) and (d) Magnified HRTEM images corresponded to region I and II outlined by dashed line in (b).



**Fig. S12** TEM images of the graphene-wrapped ZnO quantum dots assembled tubular structure after completion of 1000 discharge/charge cycles at a current density of  $2000 \text{ mA g}^{-1}$ . (a) TEM and (b) HRTEM images. (c), (d) and (e) Magnified HRTEM images corresponded to region I, II and III outlined by dashed line in (b).



**Fig. S13** FESEM images the cycled bare ZnO electrode after (a) and (b) 2 discharge/charge cycles, (c) and 100 discharge/charge cycles, and (e) and (f) 1000 discharge/charge cycles at a current density of  $2000 \text{ mA g}^{-1}$ .



**Fig. S14** The equivalent circuit used to fit the Nyquist plots of the graphene-wrapped ZnO and bare ZnO electrodes, In this model,  $R_e$  represents the internal resistance of cells, and  $R_f$  and  $CPE_1$  are associated with the resistance and constant phase element of SEI film, respectively.  $R_{ct}$  and  $CPE_2$  depict the charge transfer resistance and constant phase element of the electrode/electrolyte interface, respectively. Meanwhile,  $Z_w$  is the Warburg impedance.

**Table S1.** Summary of the electrochemical performance of various ZnO-based anode materials for lithium-ion batteries.

Materials	Morphology	Discharged capacity (mAh g <sup>-1</sup> )	Cycles	Current density (mA g <sup>-1</sup> )	Ref.
ZnO	Hierarchical flower-like nanospheres	381	30	493.5	28
ZnO	Ultralong mesoporous nanowires	392	50	98.7	29
ZnO	Ultrathin nanotubes	386	50	494	30
ZnO	Mesoporous nanosheets	421	100	20	31
ZnO	Dandelion-like nanorod arrays	310	40	--	32
Au-ZnO	Hierarchical flower-like nanostructures	392	50	120	33
Al-ZnO	Nanoparticles	418	--	50	34
ZnO-Ag-C	Porous microspheres	729	200	100	5
ZnO/Graphene	Nanocrystals	~300	25	50	17
Al-ZnO-graphene	Aerogel composite	490	100	100	6
ZnO/MWCNT	Nanotube Nanocomposite	460	100	197.4	35
ZnO/Ketjenblack	Porous structure	538.4	100	100	36
ZnO/Graphene	Nanotubes	1058	200	100	Our work
		747	200	200	
		683	200	500	
		891	1000	2000	

**Table S2.** Kinetic parameters of graphene-wrapped ZnO and bare ZnO electrodes before and after different galvanostatic discharging/charging cycles.

Graphene-wrapped ZnO electrodes	R <sub>ct</sub> (Ω)	Bare ZnO electrodes	R <sub>ct</sub> (Ω)
Before cycling	26.4	Before cycling	36.7
After 2 cycles	28.3		
After 50 cycles	214.7		
After 100 cycles	245.9	After 100 cycles	301.4
After 400 cycles	38.1		
After 700 cycles	33.1		
After 1000 cycles	13.9	After 1000 cycles	313.2