

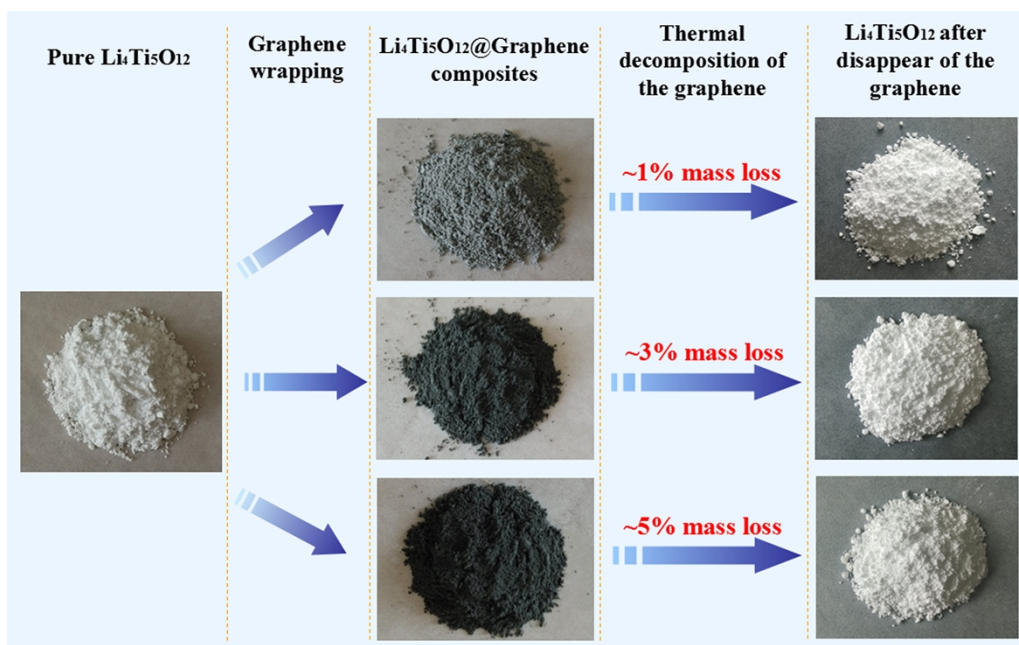
## Electronic supplementary information

### **Scalable Synthesis of Graphene-wrapped $\text{Li}_4\text{Ti}_5\text{O}_{12}$ Dandelion-like Microspheres for Lithium-ion Batteries with Excellent rate capability and Long-cycle Life**

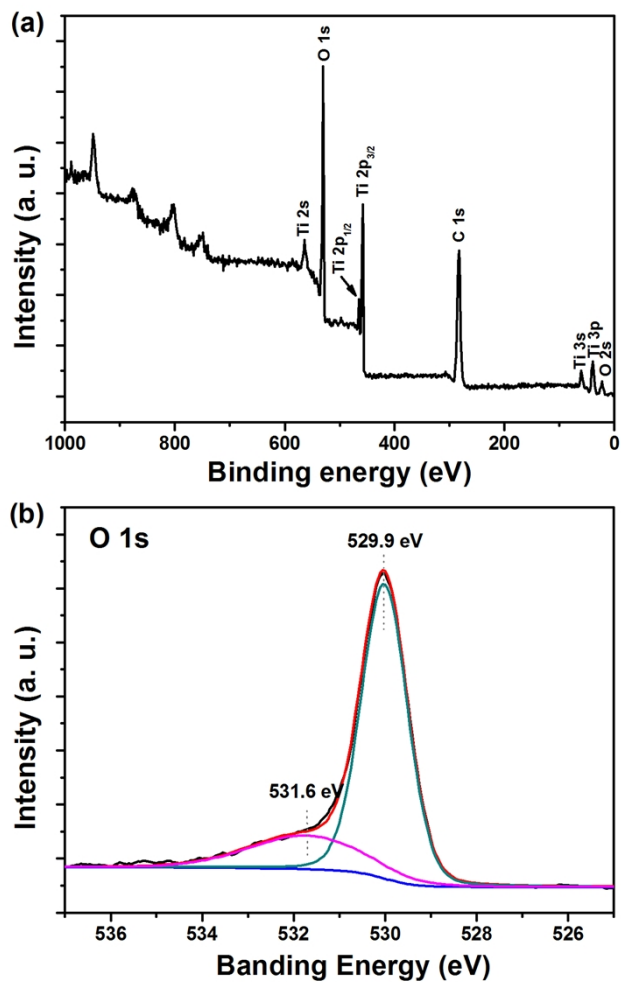
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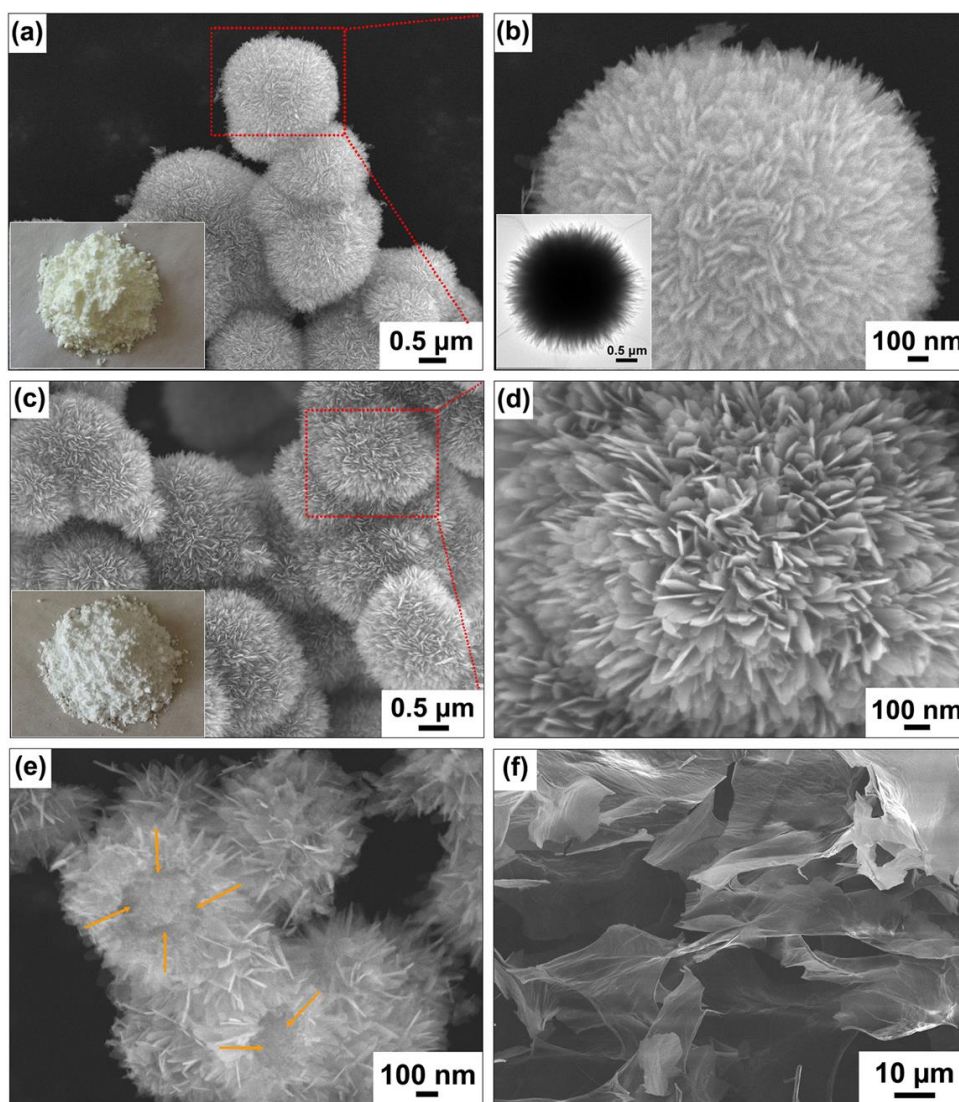
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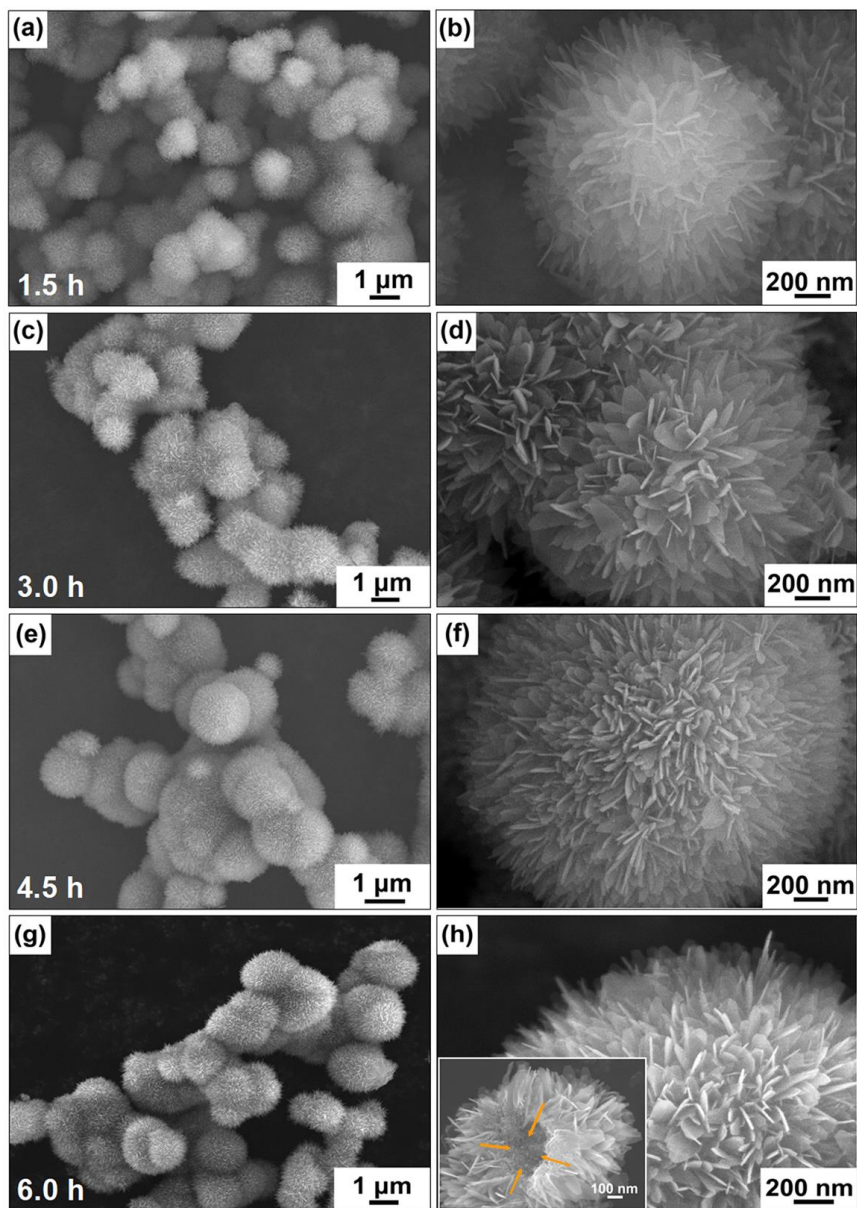
**Figure S1.** Comparison of the individual colors of the prepared samples.



**Figure S2.** (a) XPS spectra of as-synthesized  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene hierarchical heterostructures; (b) XPS survey scan of O 1s region.

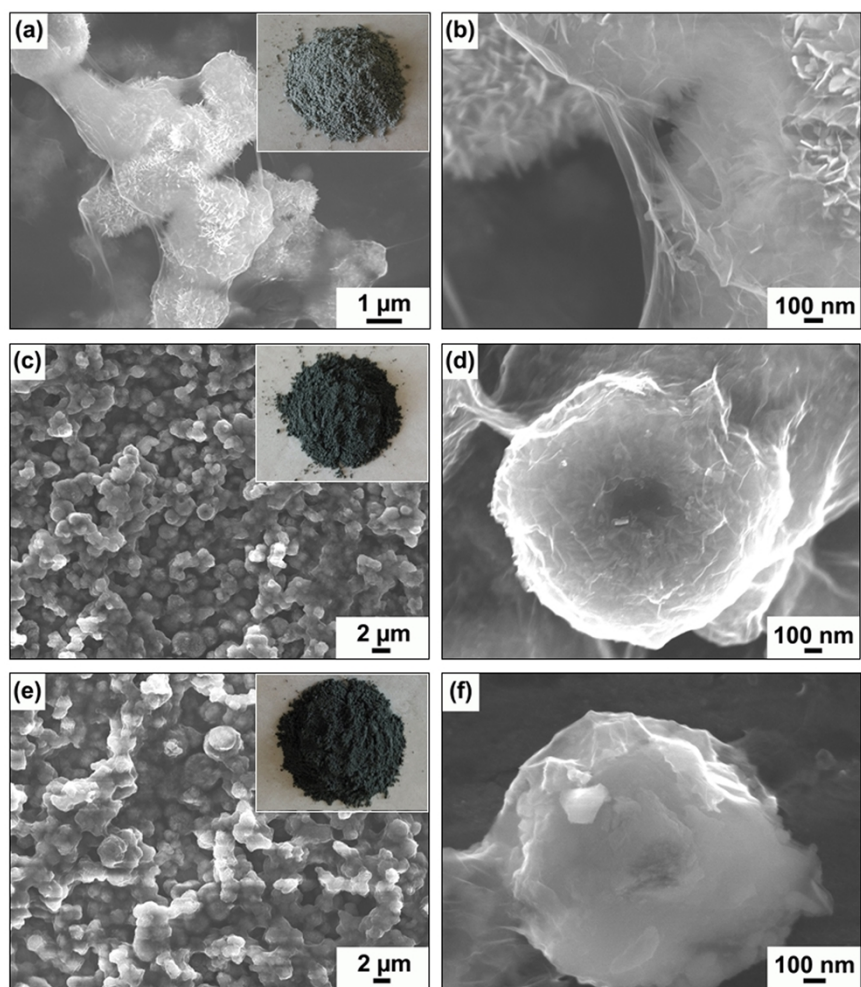


**Figure S3.** (a, b) SEM images of  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  precursor at various magnifications; (c, d) SEM images of  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres at various magnifications; (e) SEM image of a cross-sectionally damaged  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres; (f) SEM images of GO nanosheets at low-magnifications; The insets of (a) and (c) are the photographs from  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  precursor and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres, respectively, and the insets of (b) is the low-magnification TEM image of the  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  precursor.

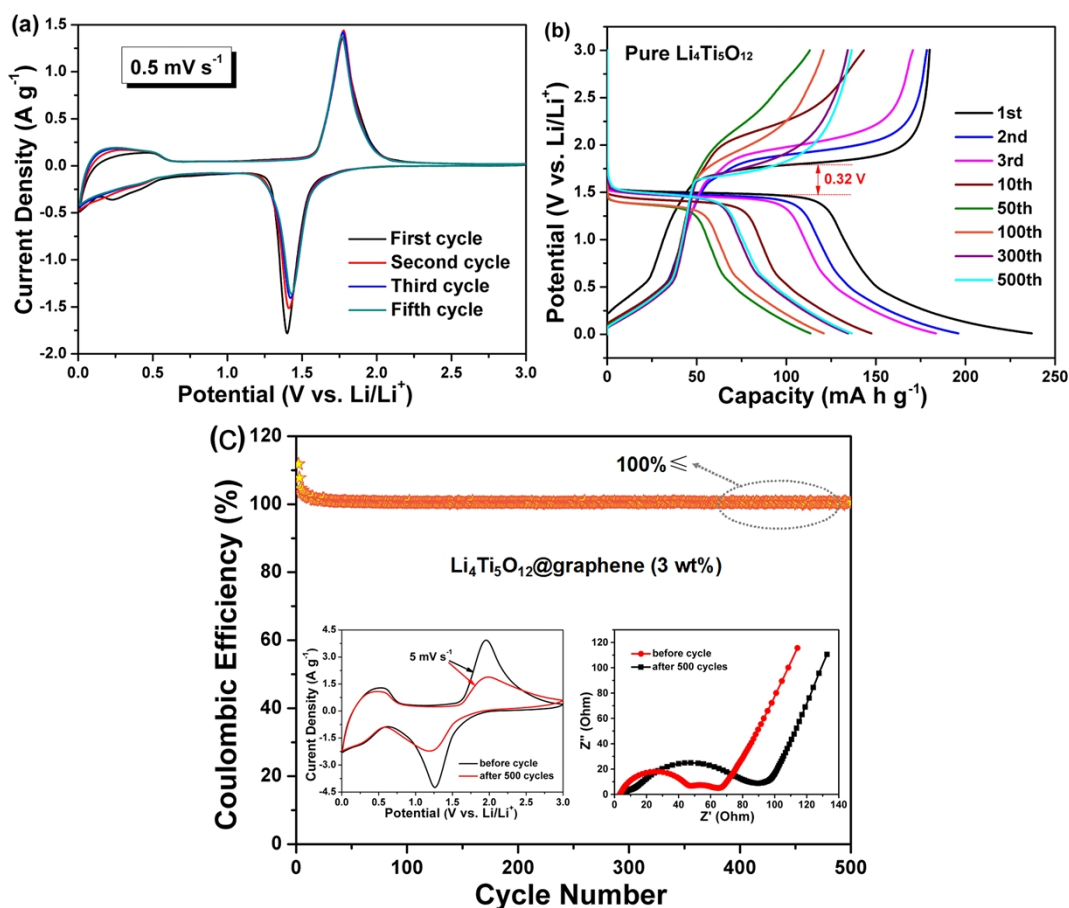


**Figure S4.** SEM images of the  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres at various reaction stages with the different reaction time: (a, b) 1.5 h, (c, d) 3.0 h, (e, f) 4.5 h and (g, h) 6.0 h. The inset of (h) is the SEM pattern from a broken  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microsphere.

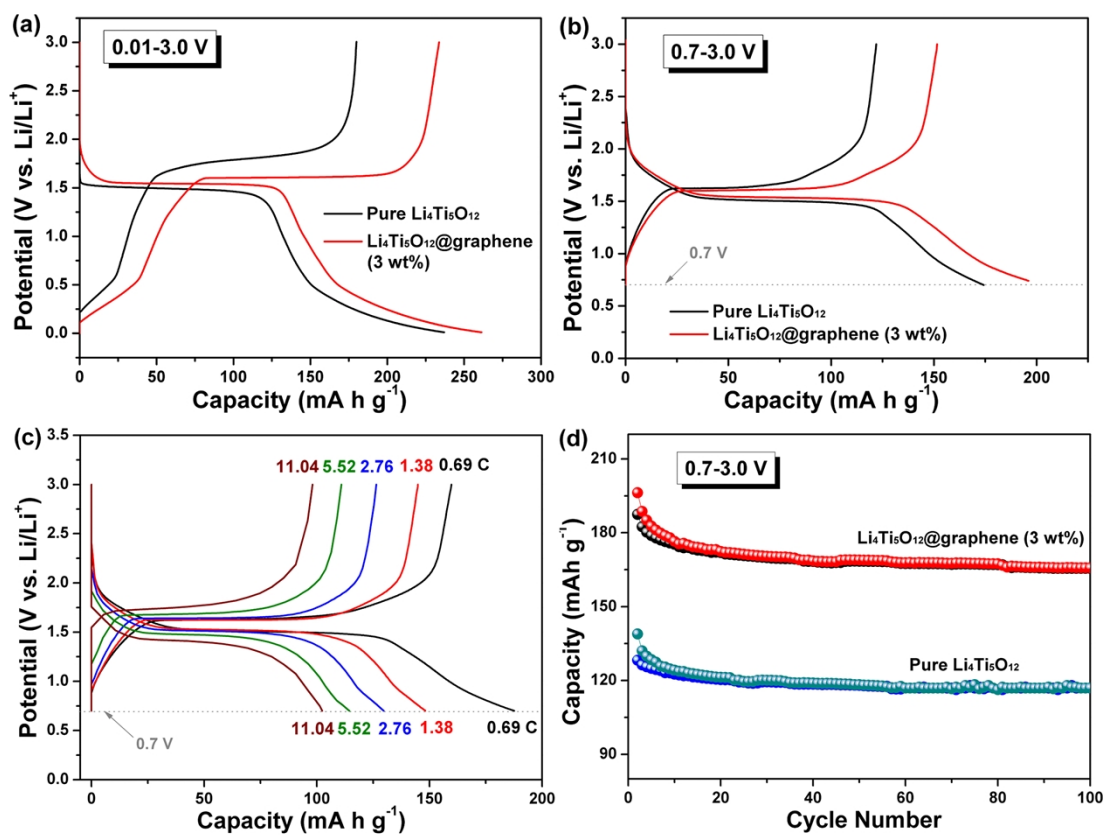




**Figure S5.** (a, b) SEM image of 1 wt% graphene coated  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres at various magnifications; (c, d) SEM image of 3 wt% graphene coated  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres at various magnifications; (e, f) SEM image of 5 wt% graphene coated  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres at various magnifications.



**Figure S6.** (a) Cyclic voltammograms of the  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene (3 wt%) between 3.0 and 0.01 V at a scan rate of  $0.5 \text{ mV s}^{-1}$ ; (b) Galvanostatic discharge/charge profiles of the pure  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  anode at a constant current density of  $0.12 \text{ A g}^{-1}$  for the 1st~500th; (c) The coulombic efficiency of 3 wt% graphene coated  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  microspheres at a charge/discharge rate of  $0.12 \text{ A g}^{-1}$ . The insets of (c) are the cyclic voltammograms (CV) curves and electrochemical impedance spectroscopy (EIS) of the  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene (3 wt%) composites electrode before and after cycling, respectively.



**Figure S7.** Initial charge-discharge curves of the pure  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene (3 wt%) composite electrodes between (a) 0.01-3.0 and (b) 0.7-3.0 V vs.  $\text{Li/Li}^+$  at 0.69 C rate; (c) Charge and discharge curves of  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene (3 wt%) discharged to 0.7 V with a different charge and discharge rate; (d) Cycling performance of pure  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene (3 wt%) discharged to 0.7 V.



**Table 1.** Specific capacitance of the  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene composites prepared in this work and of the  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  and  $\text{Li}_4\text{Ti}_5\text{O}_{12}$ -based composites reported in previous work. Information regarding the current density and the mass loading of active materials is included for better comparison among different samples.

Materials	Fabrication method	Active mass loading	Cycle potential range	Specific Capacitance	Remarks	Ref.
dandelion-like $\text{Li}_4\text{Ti}_5\text{O}_{12}$ nanospheres	hydrothermal reaction	1.42 mg/cm <sup>2</sup>	0.01-3.0 V	136 mA h g <sup>-1</sup> after 500 cycles at 0.69 C rate	~42% capacity loss for 500 cycles at 0.69 C rate	this work
			0.7-3.0 V	117 mA h g <sup>-1</sup> after 100 cycles at 0.69 C rate	~15.2% capacity loss for 100 cycles at 0.69 C rate	
$\text{Li}_4\text{Ti}_5\text{O}_{12}$ @graphene (3 wt%) composites	hydrothermal reaction	1.68 mg/cm <sup>2</sup>	0.01-3.0 V	206 mA h g <sup>-1</sup> after 500 cycles at 0.69 C rate	~21% capacity loss for 500 cycles at 0.69 C rate	this work
			0.7-3.0 V	166 mA h g <sup>-1</sup> after 100 cycles at 0.69 C rate	~15.3% capacity loss for 100 cycles at 0.69 C rate	
$\text{Li}_4\text{Ti}_5\text{O}_{12}$	solid-state method	N. A.	0.01-2.5 V	213 mA h g <sup>-1</sup> after 152 cycles at 1 C rate	~22% capacity loss for 152 cycles at 1 C rate	[1]
$\text{Li}_4\text{Ti}_5\text{O}_{12}$	solid-state method	N. A.	0-2.5 V	197 mA h g <sup>-1</sup> after 50 cycles	~19% capacity loss for 50 cycles	[2]
			0.5-2.5 V	165 mA h g <sup>-1</sup> after 50 cycles	~8.4% capacity loss for 50 cycles	
lanthanum-modified $\text{Li}_4\text{Ti}_5\text{O}_{12}$	solid-state method	N. A.	0-3.0 V	206 and 197 mA h g <sup>-1</sup> after 100 cycles at 1 C and 3 C	~3% capacity loss for 100 cycles at 1 C rate	[3]
mesoporous $\text{Li}_4\text{Ti}_5\text{O}_{12}$ hollow spheres	templating method	N. A.	1.0-3.0 V	150 mA h g <sup>-1</sup> at 2C rate	~12% capacity loss for 100 cycles at 5 C rate	[4]
hollow $\text{Li}_4\text{Ti}_5\text{O}_{12}$ urchin-like microspheres	versatile sol-gel process, hydrothermal reaction	N. A.	1.0-3.0 V	157.1 mA h g <sup>-1</sup> after 100 cycles at 2 C rate	~1.9% capacity loss for 100 cycles at 2 C rate	[5]
$\text{Li}_4\text{Ti}_5\text{O}_{12}$ -graphene hybrid nanostructures	hydrothermal reaction	N. A.	0.9-2.5V	168.5mA h g <sup>-1</sup> at 0.1 C rate	capacity loss was 3.1% at 1 C, 2.5% at 10 C and 2.9% at 60 C rate	[6]
nanosized $\text{Li}_4\text{Ti}_5\text{O}_{12}$ /graphene hybrid materials	ball milling	N. A.	0.8-2.6 V	170 mA h g <sup>-1</sup> at 1 C rate	~5.2% capacity loss for 300 cycles at 20 C rate	[7]
microscale C- $\text{Li}_4\text{Ti}_5\text{O}_{12}$ particles	solid-state reaction	N. A.	1.0-3.0 V	165 mA h g <sup>-1</sup> at 1 C rate	<1% capacity loss after 100 cycles at 1 C rate	[8]
$\text{Li}_4\text{Ti}_5\text{O}_{12}$ @C hollow microspheres	solution-phase selfassembly and subsequent solid-phase lithiation	N. A.	1.0-3.0 V	152 mA h g <sup>-1</sup> and 116 mA h g <sup>-1</sup> at 1 C and 5 C, respectively	a capacity retention of 120 mA h g <sup>-1</sup> after 80 cycles at 10 C rate	[9]
$\text{Li}_4\text{Ti}_5\text{O}_{12}$ /C composite	hydrothermal method	N. A.	1.0-2.5 V	176 mA h g <sup>-1</sup> and 136 mA h g <sup>-1</sup> at 0.1 C and 20 C, respectively	~19.7% and 24.2% capacity loss for 200 cycles at 10 and 20C rate	[10]
$\text{Li}_4\text{Ti}_5\text{O}_{12}$ nanosheets/ N-doped carbon	N. A.	N. A.	0.9-2.5V	170.1 mA h g <sup>-1</sup> at 1C rate	<1% capacity loss after 100 cycles at 10 C rate	[11]
Microscale C- $\text{Li}_4\text{Ti}_5\text{O}_{12}$ particles	Solid-state reaction	N. A.	1.0-3.0 V	165 mA h g <sup>-1</sup> and 123 mA h g <sup>-1</sup> at 1 C and 10 C, respectively	~6.3% capacity loss for 100 cycles at 10 C rate	[12]

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