# Synthesis and electrochemical properties of Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub>/g-C<sub>3</sub>N<sub>4</sub> composites as anode materials for Li-ion batteries

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#### 1. Synthesis of Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub>/g-C<sub>3</sub>N<sub>4</sub> composite

First, g-C<sub>3</sub>N<sub>4</sub> light yellow powder was obtained by calcining 6 g melamine in air at 500 °C for 2 h at 10 °C /min. Then, 3.2926 g Zn(CH<sub>3</sub>COO)<sub>2</sub>·2H<sub>2</sub>O was put into the mixed solvent of ethanol and glycol(volume ratio 1:1), and the clarified solution was obtained by magnetic stirring. 5.1045 g tetrabutyl titanate and ethanol solutions with different proportions of g-C<sub>3</sub>N<sub>4</sub> were added to the solution and then transferred to a polytetrafluoroethylene reactor, which was heated at 180 °C for 10 h. The products of solvothermal reaction were filtered, washed 3 times by ethanol and dried at 70 °C for 5h. The product without g-C<sub>3</sub>N<sub>4</sub> was calcined in air at 500 °C for 2 h, and pure Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub> are acquired. Then others were calcined in a tubular furnace at 600 °C for 2 h under the protection of N<sub>2</sub>, whereafter calcined in air at 500 °C for 2 h. Finally, the Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub>/g-C<sub>3</sub>N<sub>4</sub> (1, 3 and 8 wt%) composites were triumphantly obtained.

### 2. Materials characterization

The composite  $Zn_2Ti_3O_8/g-C_3N_4$  (0, 1, 3 and 8 wt%) composites were measured by X-ray diffraction (XRD, Shimadzu XRD-6100). The  $Zn_2Ti_3O_8/g-C_3N_4$  (0, 1, 3 and 8 wt%) composites were represented via X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific Escalab 25Xi). The morphologies of these composites were analyzed by scanning electron microscopy (SEM, JSM-7800F Prime), transmission electron microscopy (TEM) and selected area electron diffraction (SAED, JEM-2010).

#### 3. Electrochemical measurements

The electrochemical performance of Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub>/g-C<sub>3</sub>N<sub>4</sub> (0, 1, 3 and 8 wt%) anode

materials were studied using the CR2032 cells. The obtained anode material, dry acetylene black and polyvinylidene fluoride were evenly mixed according to the mass ratio of 7:2:1. And 1-methyl-2-pyrrolidone was dripped in a suitable amount and then grinded adequately. It is evenly coated on the copper foil in a dry environment. Afterwards, the anode electrode was acquired via drying at 105°C for 12 h in a vacuum drying oven. Copper foils placed on the slicer were cut into sheet electrodes, then weighed and recorded. Finally, the batteries were assembled in a glove box filled with high purity Ar. In brief, Lithium plate was used as counter electrode, Cegard 2400 porous polymer membrane was used as separator, and 1 mol·L<sup>-1</sup> LiPF<sub>6</sub>/EC-DMC mixed solution (volume ratio 1:1) was used as electrolyte. Constant current charge/discharge measurements were carried out on CT2001A instrument at different current rates of 50, 100, 300, 600, 900 and 1500 mA $\cdot$ g<sup>-1</sup> in the potential range of 0-3V. According to the weight of  $Zn_2Ti_3O_8/g$ -C<sub>3</sub>N<sub>4</sub> (0, 1, 3 and 8 wt%) composites, the charge / discharge capacity of the electrode was calculated. The electrochemical impedance spectroscopy (EIS) of the anode materials was carried out at partat 4000 electrochemical workstation of AMETEK company in the frequency range of  $10^{5}$ - $10^{-2}$  Hz

#### 2. Results and discussion



Fig. S1. The used refinement model of Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub>.



Fig. S2. Rietveld refinements of (a) pure  $Zn_2Ti_3O_8$ , (b)  $Zn_2Ti_3O_8/g$ -C<sub>3</sub>N<sub>4</sub> (1 wt%),

(c)  $Zn_2Ti_3O_8/g$ -C<sub>3</sub>N<sub>4</sub> (3 wt%) and (d)  $Zn_2Ti_3O_8/g$ -C<sub>3</sub>N<sub>4</sub> (8 wt%).