Electronic Supplementary Information (ESI)

An internal magnetic field strategy to reuse pulverized active materials for high performance: magnetic three-dimensional ordered macroporous TiO_2/CoPt/α-Fe_2O_3 nanocomposite anode

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1. Experimental

All chemicals were analytical grade and purchased from Aladdin Chemicals. The fabrication process of 3DOMTCF and 3DOMTC nanocomposites are composed of colloidal crystal template and sol-gel methods, according to previous literatures[1-2].

1.1 Preparation of poly methyl methacrylate (PMMA) microspheres

66g methyl methacrylate was added to 260 ml deionized water .The solution was stirred until the temperature was 75 ℃.Then, 0.25g AIBA was added to this solution and the mixture was stirred for 2h at the temperature of 75 ℃.

1.2 Preparation of PMMA colloidal crystal template

30ml as-prepared PMMA solution was vigorously agitated using an ultrasound cleaner for 10 min three times. Then the solution was dried at 60℃ for 3~4h.

1.3 Preparation of 3DOM TiO_2

The precursor was prepared by adding 1.0ml hydrochloric acid, 5.0ml titanium isopropoxide, 2.0ml deionized water into 5.0ml ethanol, successively. The precursor and PMMA colloidal crystal template was mixed in a mass rate of 1:1. After vacuum

suction filtration, the mixture was dried at room temperature for 24h. Then the mixture was annealed in air atmosphere at 300 ℃ for 2h (heating rate of 2℃/min), subsequently, calcined at 500 ℃ for 2h (heating rate of 2℃/min).

1.4 Preparation of 3DOMTC

0.68 g Cobalt nitrate($Co(NO_3)_2 \cdot 6H_2O$) and 1g Chloroplatinic acid（$H_2PtCl_6 \cdot 6H_2O$）were dissolved in 2ml deionized water, respectively. Then the resultant solutions were mixed. After stirring for 1h, 4ml Citric acid solution (0.98g Citric acid) was added into the mixture. The result solution was reserved at room temperature for 24h.

3DOM TiO_2 mixed with appropriate the pre-prepared solution. after vigorous agitation using an ultrasound cleaner for 1.5h, the mixture was dried at 60℃ for 24h. 3DOMTC was synthesized by annealing the mixture in Ar atmosphere at 700 ℃ for 1h (heating rate of 1℃/min).

1.5 Preparation of 3DOMTCF

0.2mol $FeCl_2 \cdot H_2O$,0.2mol Citric acid and 8g ascorbic acid were dissolved in 200ml ethanol. After ring for 6h at 60℃, 0.1mol DMF was added into the pre-prepared. Subsequently, the solution continued to be stirred for 30min at 60℃. Appropriate 3DOMTC composites were added into the pre-prepared solution, and dried over night at 100℃. 3DOMTCF synthesized by annealing the mixture in Ar atmosphere at 700 ℃ for 1h (heating rate of 1℃/min).

The content of α-Fe_2O_3, CoPt and TiO_2 in the final product of 3DOMTCF was 47 wt.%, 33.7 wt.%, and 19.3 wt.% respectively, which was determined through ICP-MS.

1.6 Physical Characterizations

The power X-ray diffraction (XRD) measurement was taken by an XRD diffractometer (X' Pert Pro, PANalytical, Netherlands) using Cu Ka radiation (λ=1.546Å). The morphology, energy-dispersive X-ray spectroscopy (EDX) mapping of the resultant composites were obtained by a field emission scanning electron microscope (FESEM) (Hitachi S-4700, Hitachi, Japan) and transmission electron microscopy (TEM) (Tecnai G2F30 S-TWIN, PANalytical, Netherlands). Hysteresis loop was determined by NQTM-DC type vibrating sample magnetometer test analysis. The test temperature was room temperature (25 ℃), and the magnetic field ranged from-1.25 to 1.25Tesla.

1.7 Electrochemical Measurements

The electrochemical properties were obtained from CR2032-type coin cells. Active materials, conductivity agent (carbon black), and binder (polyvinylidene fluoride, PVDF) were added in turn with a weight ratio of 8:1:1 into a beaker. Subsequently, the slurry was obtained by compressing onto a copper foil and vacuum dried at 120 °C for 10h to fabricate working electrodes. The magnetized 3DOMTCF electrodes were magnetized by using magnet. The electrolyte consisted of LiPF6 (1 M) in a 1:1 (v/v) mixture of ethylene carbonate (EC)/diethyl carbonate (DEC). Coin cells were assembled in an argon-filled glove box and the H_2O and O_2 content was below 1

and 0.1ppm, respectively. After 24h aging treatment ,the galvanostatic charge/discharge tests were carried out on a Land CT2001A battery testing system (Wuhan, China) at different rate with the potential ranging from 0.01 to 3.00 V (vs. Li/Li^+). Cyclic voltammetry (CV) measurement was performed on a CHI760D electrochemical workstation(Shanghai, China) at a scan rate of 0.1 mV s^{-1}, with the potential range of 0.01to 3.00 V (vs. Li/Li^+).

2. Results

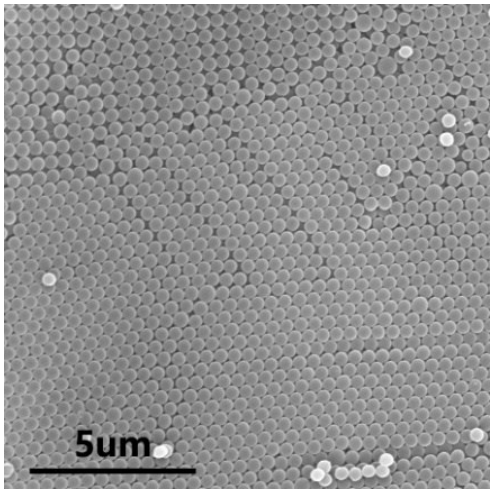

Figure S1. SEM image of PMMA colloidal crystal template.

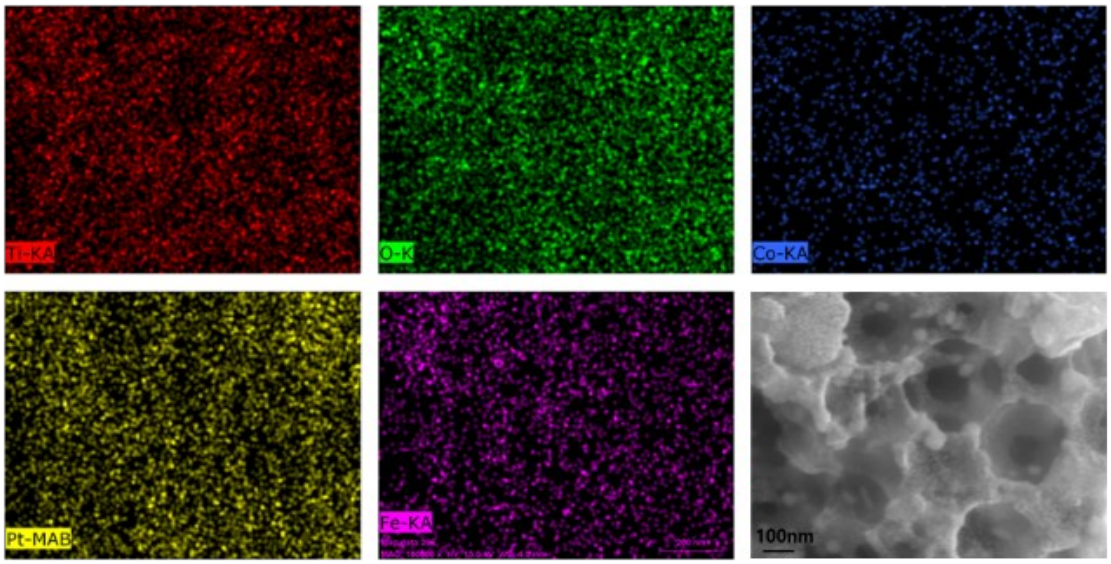

Figure S2. EDX elemental mappings of 3DOMTCF.

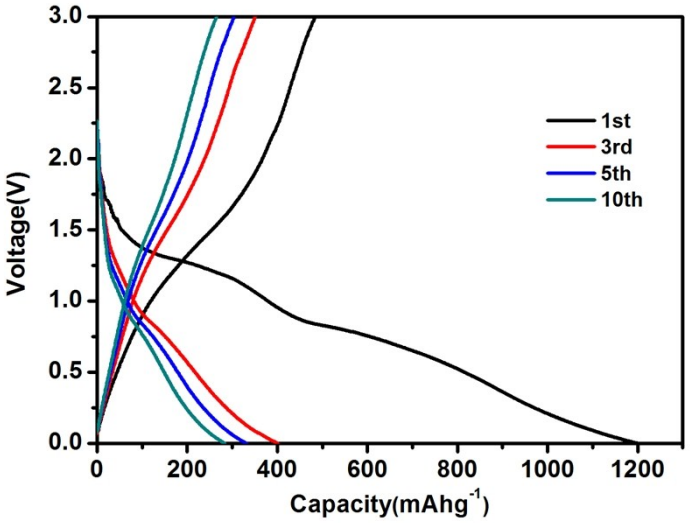

Figure S3. Charge- discharge profiles of the unmagnetized 3DOMTCF with a voltage window of 0.01-3.00V at a current density of 50 mA/g

Table S1. Saturation magnetization, coercirity and remanent magnetization of different samples.

Sample	Temperature [°C]	Saturation magnetization(Ms) [emu/g]	Coercivity(HC) [Oe]	Remanent magnetization(MR) [emu/g]
3DOMTC	25	20.39	817.54	7.63
3DOMTCF	25	9.39	913.61	3.60

References

1 B.T. Holland, C. F. Blanford, T. Do, A. Stein, Chem. Mater., 1999, **11**, 795-805.

2 A. Vu, Y. Qian, A. Stein, Adv. Energy Mater., 2012, **2**, 1056-1085.