Supporting Information

Pyrene-4,5,9,10-tetraone/Graphene Composite Film as a Sustainable Cathode Material for Aqueous Iron-Ion Batteries

Wenwen Deng[a,b,#], Ning Zhang[c,#], Ying Zhou[b], Yuanyuan Quan[b], Lei Yao[b], Chengyang Zhang[b], Yongxin Huang[c*], Zhong Jin[a*]

a State Key Laboratory of Coordination Chemistry, MOE Key Laboratory of Mesoscopic Chemistry, MOE Key Laboratory of High Performance Polymer Materials and Technology, Jiangsu Key Laboratory of Green Energy Catalysis and Intelligent Chemical Engineering, Suzhou Key Laboratory of Green Intelligent Manufacturing of New Energy Materials and Devices, Tianchang New Materials and Energy Technologies Research Center, Institute of Green Chemistry and Engineering, School of Chemistry and Chemical Engineering, Nanjing University, Nanjing, Jiangsu 210023, China.

b School of Material Science and Engineering, Suzhou University of Science and Technology, Suzhou 215000, China

c Beijing Key Laboratory of Environmental Science and Engineering, School of Material Science and Engineering, Beijing Institute of Technology, Beijing 100081 China

Experimental

Materials Synthesis

Pyrene-4,5,9,10-tetraone (PTO, 98%, Aladdin Co., Ltd) was used as received. PTO/graphene composites were prepared by dispersing PTO with graphene at various mass ratios (0.1 wt%, 0.3 wt%, 0.5 wt%, and 0.7 wt%) in 10 mL of ethanol under ultrasonic treatment for 30 minutes. Subsequently, 50 mL of deionized water was added to the resulting solution as an antisolvent. The mixture was then filtered under vacuum to obtain PTO and PTO/graphene composite films.

Materials Characterization

X-ray diffraction (XRD) patterns were recorded using a Panalytical X'pert PRO MRD diffractometer (Holland) with Cu Kα radiation. The morphology and particle size of the materials were examined using a field-emission scanning electron microscope (FE-SEM, JSM-7600, JEOL, Japan). Fourier-transform infrared (FTIR) spectra were collected using a Bruker Vertex 70 spectrometer. X-ray photoelectron spectroscopy (XPS) analyses were performed on a VG MultiLab 2000 system equipped with a monochromatic Al Kα X-ray source (Thermo VG Scientific).

Electrochemical Characterization

CR2032 coin-type cells were assembled to investigate the electrochemical performance. The PTO/graphene films were directly pressed onto titanium mesh to serve as the working electrode, with a diameter of 0.5 cm and an active material loading of approximately 1 mg cm^{-2}. A Whatman GF/D glass fiber membrane and 1 mol L^{-1} $FeSO_4$ aqueous solution were used as the separator and electrolyte, respectively. The electrolyte-to-active material (E/A) ratio was controlled at 8 μL mg^{-1}. Activated carbon was pressed onto nickel foam and used as the counter electrode. Galvanostatic charge–discharge tests were conducted on a battery testing system (Land CT2001A, China) within a voltage window of 0.1–0.95 V at room temperature. The capacities were calculated on PTO only. Cyclic voltammetry (CV) curves were obtained using a Princeton P2000 electrochemical workstation (USA) at a scan rate of 0.1 mV s^{-1} in the same voltage range. The pouch cells were assembled using the same PTO/graphene composite films as the cathode and activated carbon as the counter electrode. A glass fiber membrane was employed as the separator, and a 1 mol L^{-1} $FeSO_4$ aqueous solution was used as the electrolyte. The effective electrode area of the pouch cell was approximately 1 × 3 cm^2, with a cathode mass loading of about 2.5 mg cm^{-2}.

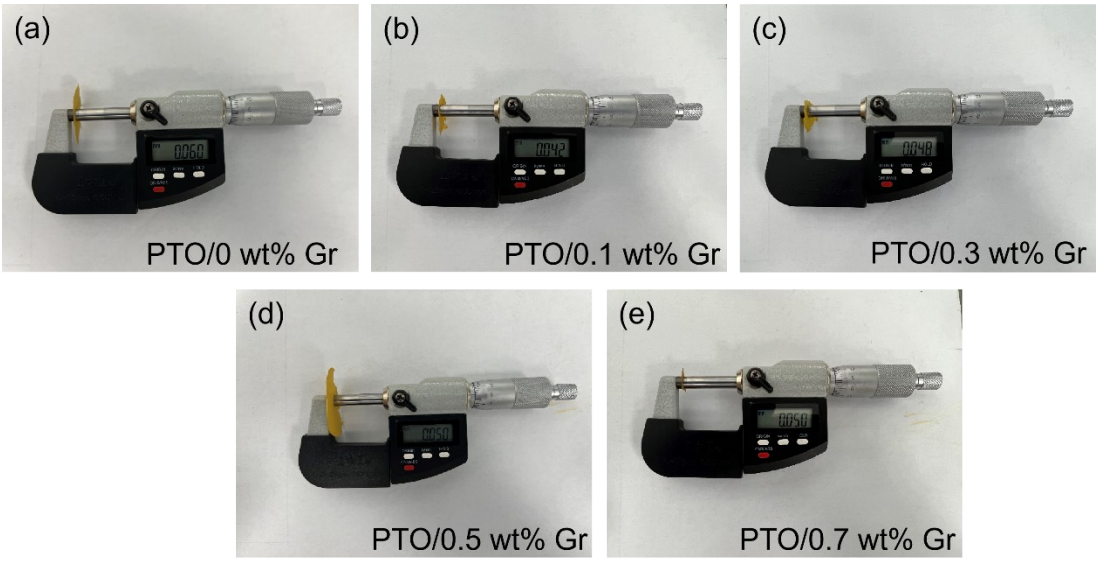

Figure S1 Images and Digital micrometer measurements of the thickness of PTO/graphene composite films with different graphene contents: (a) PTO/0 wt% Gr, (b) PTO/0.1 wt% Gr, (c) PTO/0.3 wt% Gr, (d) PTO/0.5 wt% Gr, and (e) PTO/0.7 wt% Gr.

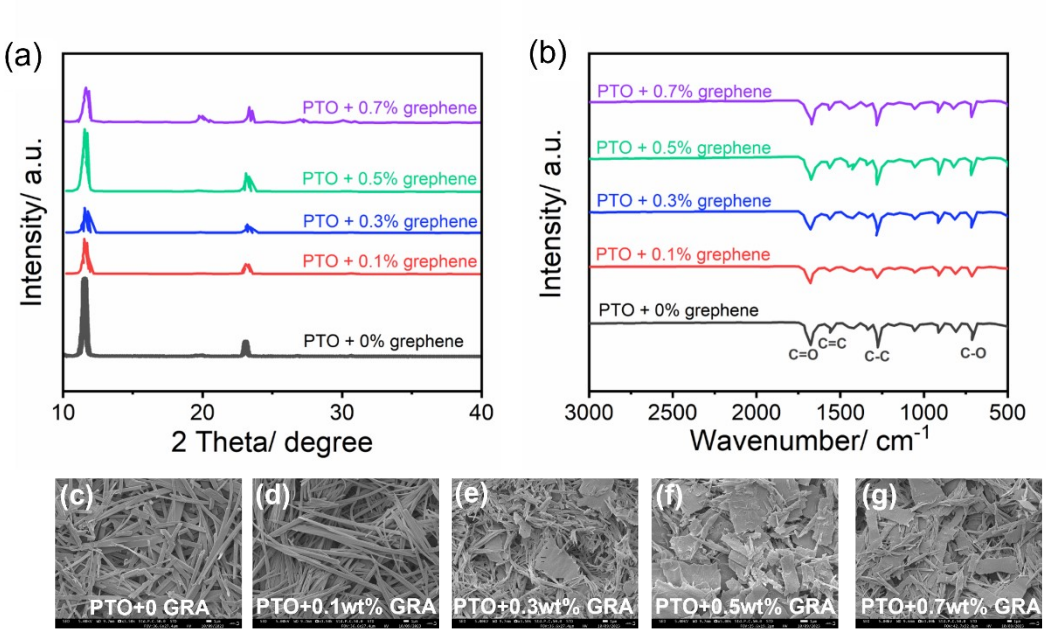

Figure S2 (a) X-ray diffraction (XRD) patterns of PTO/graphene composites with different graphene contents (0–0.7 wt%); (b) Fourier transform infrared (FTIR) spectra

of PTO/graphene composites with varying graphene loadings, showing the characteristic vibrational bands of PTO; (c–g) Scanning electron microscopy (SEM) images of PTO/graphene composite electrodes with graphene contents of (c) 0 wt%, (d) 0.1 wt%, (e) 0.3 wt%, (f) 0.5 wt%, and (g) 0.7 wt%, respectively.

Table S1 Electrochemical performance comparison of representative cathode materials for aqueous iron-ion batteries

Cathode	Specific Capacity ($mAh\ g^{-1}$)	Capacity Retention	References
PANi	75@ 200 $mA\ g^{-1}$	96% after 200 cycles	[1]
VO_2	170@ 500 $mA\ g^{-1}$	80% after 200 cycles	[2]
$VOPO_4{\cdot}2H_2O$	105@100 $mA\ g^{-1}$	80% after 50 cycles	[3]
PTO/Gr	220@ 200 $mA\ g^{-1}$	83% after 200 cycles	This work

References

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