Supporting information

One-pot synthesis and versatile applications of recyclable aminal-linked dynamic framework

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1. Experimental

1.1 Materials

CNTs were provided from Shenzhen NANO Tech. Port. Co. Ltd. The *ac*ryl chloride-functionalized CNT (CNT-$SOCl_2$) was prepared according to the literature.[1] Nitric acid (A.R.), sulfuric acid (A.R.), alcohol (A.R.), p-Phenylenediamine (PDA), paraformaldehyde, N-Methyl-2-pyrrolidone (NMP) were purchased from Aldrich and used directly. Thionyl chloride (A.R.) and N,N-Dimethylformamide (DMF) (A.R.) were used after removing the water.

1.2 Synthesis of PHDN and PHDN-CNTs nanocomposites

The fabrication of PHDN-CNT containing 2 wt% CNTs (named M-2) was used as an example to illustrate the synthetic procedure. CNT-$SOCl_2$ (0.0080 g), PDA (1.00 mmol), and paraformaldehyde (5.0 equiv, 5.0 mmol) were put into 3 mL of NMP, and then sonicated and stirred for 1 h, and reacted at 50 ^oC for 24h under N_2 atmosphere. After reaction finished, the solution was dumped into acetone, then filtered and dried overnight. Yield: 0.2340 g, dark yellow. M-0 (pure PHDN), M-0.5 (containing 0.5 wt% CNTs), M-1 (containing 1 wt% CNTs) and M-5 (containing 5 wt% CNTs) were prepared by an analogous procedure.

1.3 Recyclable Treatment

50 mg PHDN and PHDN-CNT composites are dispersed into 50 mL of 0.5 M H_2SO_4 (pH = 0) for 0.5h until degraded completely. Then the mixture solution was neutralized to pH 6.5~7.0. The precipitate was filtered and collected.

1.4 Measurements and Characterization

SEM measurement was carried out on an S-4800 instrument; FT-IR spectra were performed on a PerkinElmer Spectrum 100 Model FT-IR spectrometer in the range of 4000-400 cm^{-1}; Raman spectra were recorded on a micro-Raman spectrometerr (T64000 Jobin Yvon); XRD analysis was conducted by a D/max-γBX X-ray; TGA and DSC was conducted with PerkinElmer Pyris in flowing N_2 at a heating rate of 10 °C /min; XPS analysis was performed on a VG ESCALABMK II; The Young's modulus and hardness of films were measured by Nano-Indenter XP system.

1.5 Electrochemical property

The electrochemical properties were obtained via CHI 604B electrochemical workstation. The PHDN and the composites M5 were carbonized at 700 °C under N_2 atmosphere for 2 h, the obtained active materials are named as a-PHDN and a-M5, respectively. A mixture of active material (a-PHDN or a-M5), carbon black, and polytetrafluoroethylene (PTFE) at a weight ratio of 8:1:1 was pasted onto nickel foam (6 M KOH electrolyte) as the working electrode, the Pt wire and Hg/HgO electrode were acted as the counter electrode and reference electrode in 6 M KOH. The mass load of active material was about 6 mg cm^{-2}.

The specific capacitances are calculated using the following Eq.:

$$C_S = I \times \Delta t / (m \times \Delta V)$$

where C_S is the specific capacitance (F g^{-1}), I is the constant discharge current (A), m is the mass of active materials in the electrode (g), and Δt is the discharge time (s) in the potential window ΔV (V).

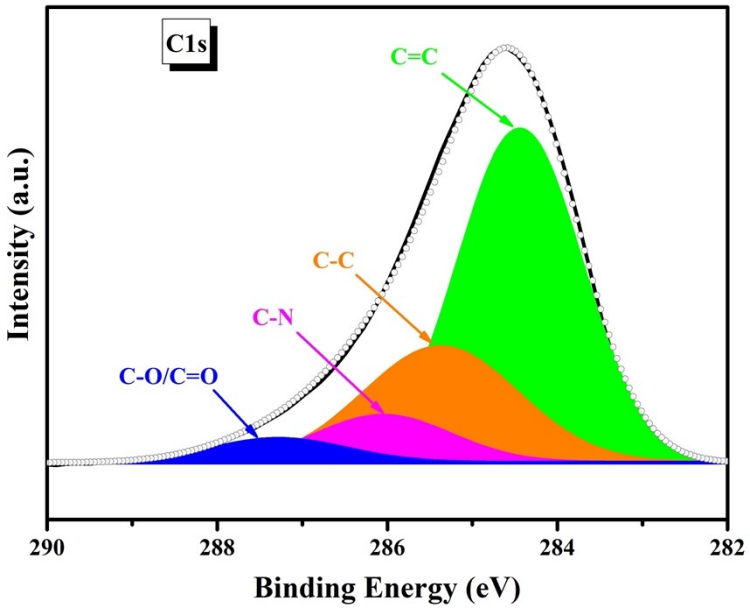

Fig. S1 Deconvoluted XPS spectra of C1s.

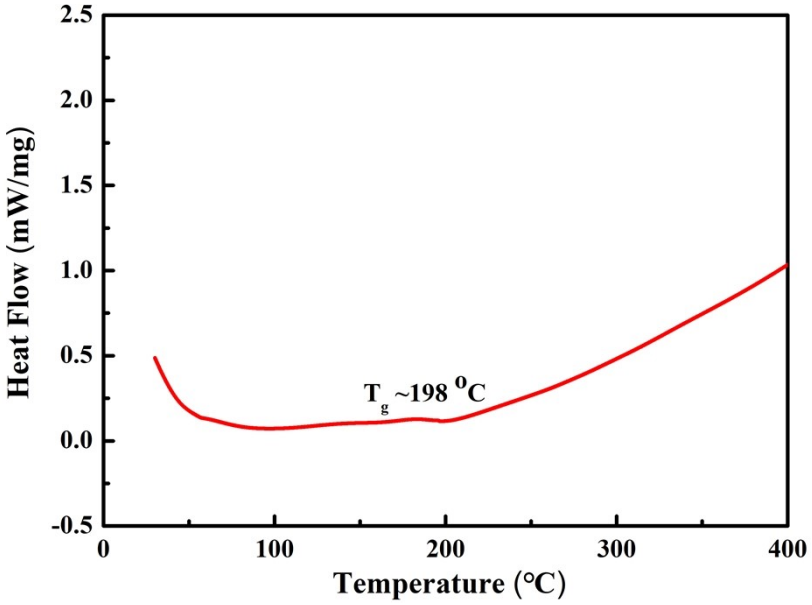

Fig. S2 DSC curves of the PHDN measured in N_2. Heating rate: 10 °C/min.

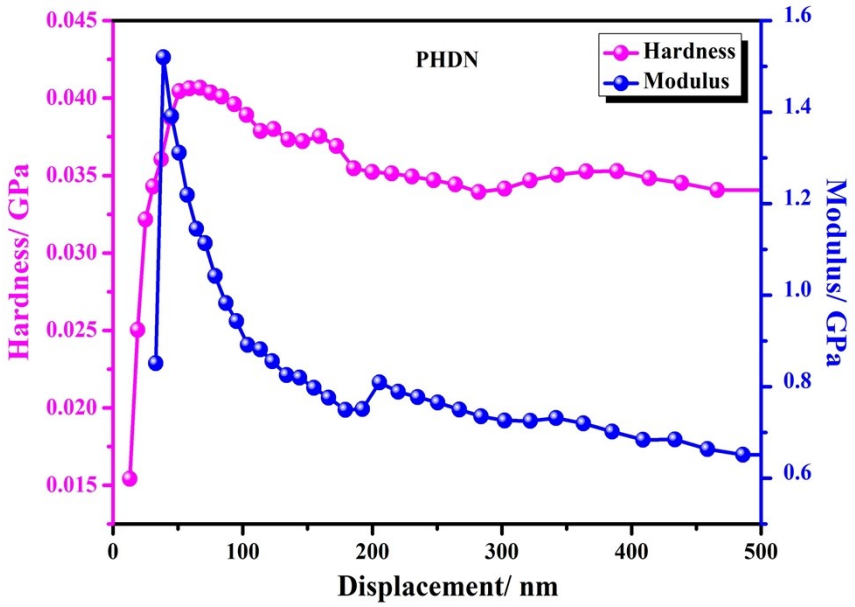

Fig. S3 Hardness and modulus curves for PHDN films.

Table S1 Decomposition rate of PHDN and the composites.

Samples	pH = 2 (RT)	pH = 0 (RT)
PHDN	~45 min decomp	< 30 min decomp
M-0.5	80 %	—
M-1	77 %	—
M-2	72 %	—
M-5	68 %	—

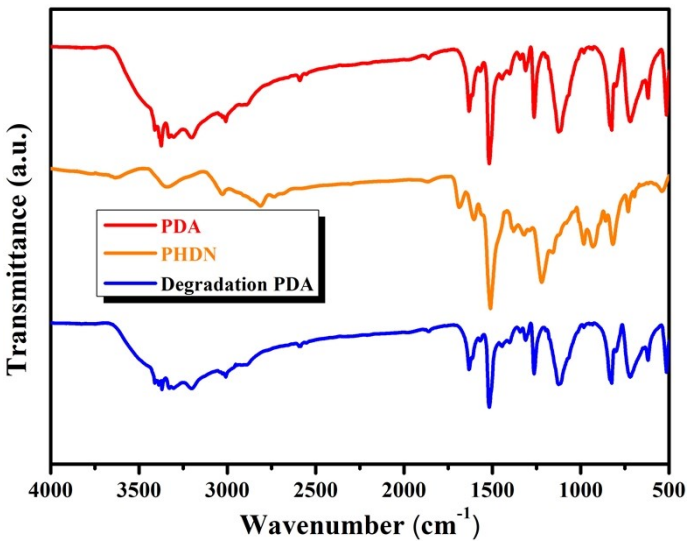

Fig. S4. IR spectra of PHDN, PDA and the degradation.

Reference

1. J. M. Garcia, G. O. Jones, K. Virwani, B. D. McCloskey, D. J. Boday, G. M. ter Huurne, H. W. Horn, D. J. Coady, A. M. Bintaleb, A. M. S. Alabdulrahman, F. Alsewailem, H. A. A. Almegren and J. L. Hedrick, *Science*, 2014, **344**, 732-735.