Supporting Information

Redox Additive-improved Electrochemically and Structurally Robust

Binder-free Nickel Pyrophosphate Nanorods as Superior Cathode for

^aNano-Electro Mechanical Device Laboratory, School of Mechanical Engineering, Yonsei

Hybrid Supercapacitors

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S1

Equations

The specific capacitance (C_g) , and specific capacity (C_{gc}) are calculated using the following equation

$$C_g = \frac{I \times \Delta t}{m \times \Delta V} \quad F g^{-1}$$
 (S1)

$$C_{gc} = \frac{I \times \Delta t}{m} \quad C g^{-1}$$
 (S2)

Mass balancing

As is known, the mass balancing between the positive and negative electrode is essential to achieve high performance supercapacitor. The mass balancing is calculated using the following equation as

$$\frac{m_{-}}{m_{+}} = \frac{Q_{+}}{C_{-} \times \Delta V_{-}} \tag{S3}$$

Where, m_+ & m_- is active material loading in positive and negative electrodes respectively (mg), C_- is the specific capacitance of after pyrolysis graphene (F g^{-1}), and ΔV_- is the potential window (V). According to the above-mentioned equation, the calculated mass ratio between negative and positive electrode is 5.5.

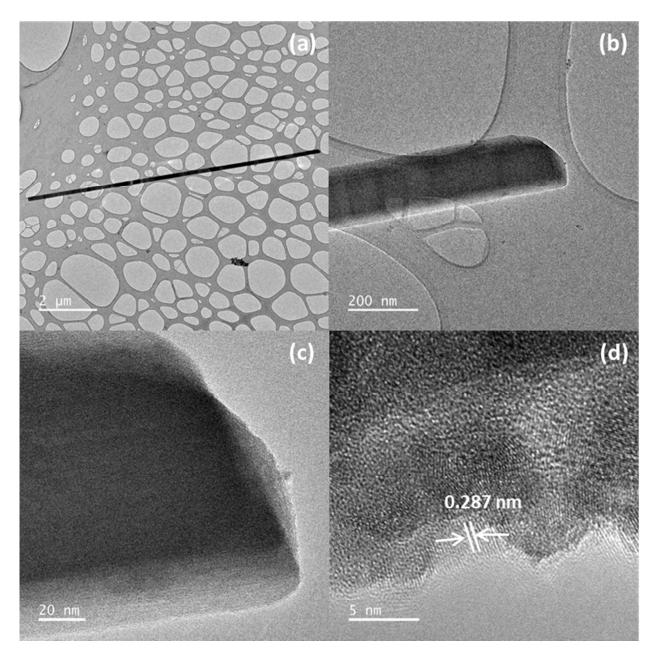


Fig. S1. (a-c) TEM images of the nickel pyrophosphate 1D nanorods at different magnifications.

(d) HRTEM image of the nickel pyrophosphate 1D nanorods.

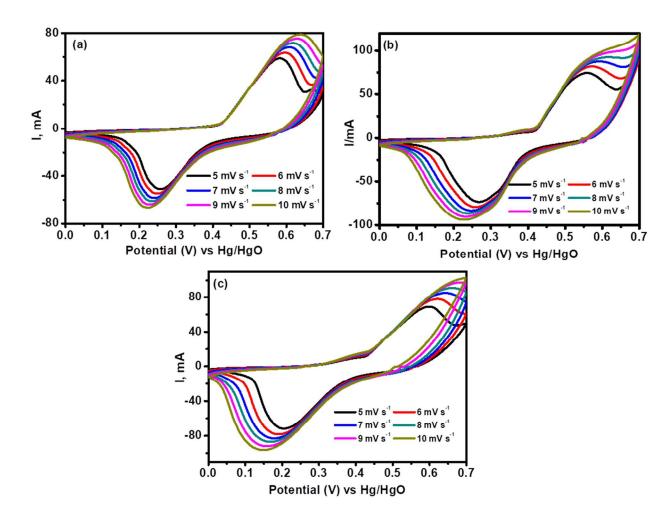


Fig. S2 CV curves of nickel pyrophosphate 1D nanorods at various scan rates in (a) 1M KOH, (b) 1M KOH+0.050mg $K_3[Fe(CN)_6]$ and (c) 1M KOH+0.100 mg $K_3[Fe(CN)_6]$ electrolytes respectively.

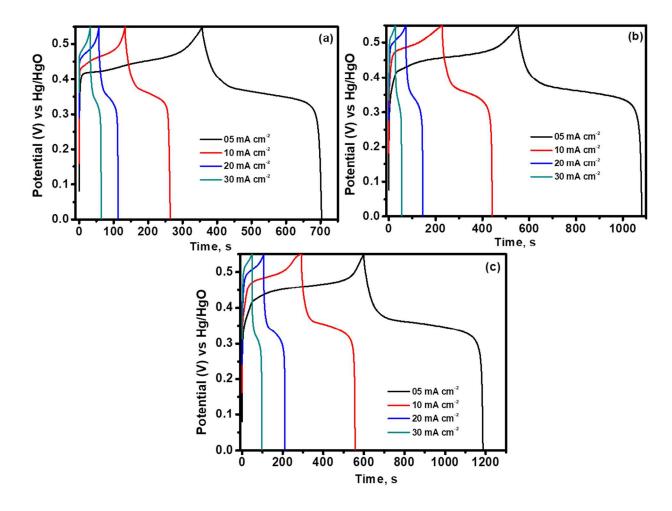


Fig. S3 GCD curves of nickel pyrophosphate 1D nanorods at various current densities in (a) 1M KOH, (b) 1M KOH+0.050mg $K_3[Fe(CN)_6]$ and (c) 1M KOH+0.100 mg $K_3[Fe(CN)_6]$ electrolytes respectively.

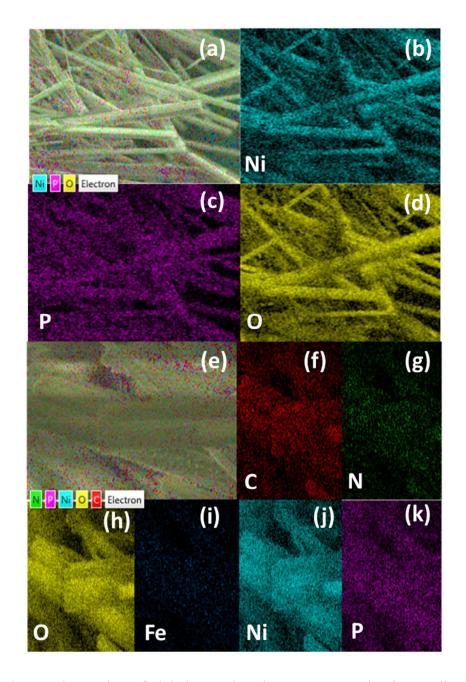


Fig. S4 The elemental mapping of nickel pyrophosphate 1D nanorods after cycling in (a-d) 1M KOH and (e-k) 1M KOH+0.075 mg $K_3[Fe(CN)_6]$ electrolytes respectively.

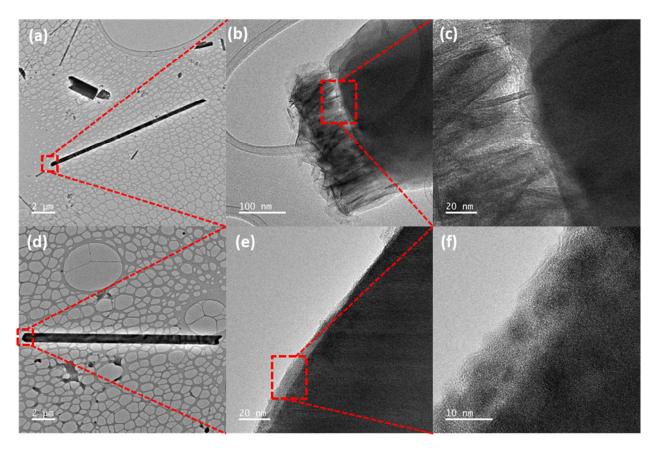


Fig. S5 TEM images of nickel pyrophosphate 1D nanorods after cycling in (a-c) 1M KOH and (d-f) 1M KOH+0.075 mg $K_3[Fe(CN)_6]$ electrolytes respectively.

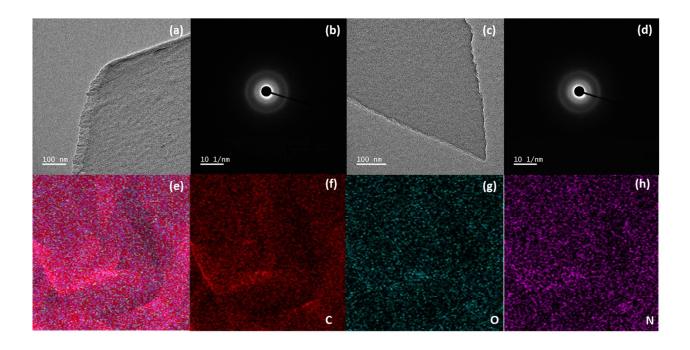


Fig. S6 TEM images and SAED pattern of N-doped rGO before (a, b) and after (c, d) pyrolysis. The representative N, O, and C elements (e-h) distribution on the graphene sheet after pyrolysis.

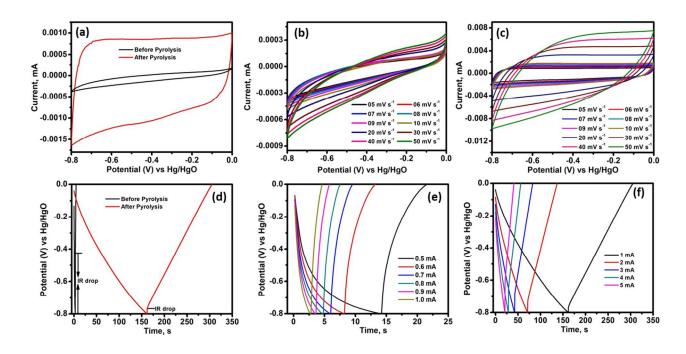


Fig. S7 (a) comparison CV curves of N-doped rGO at 5 mV s⁻¹. (b, c) CV curves of N-doped rGO at different scan rates. Comparison (d) GCD curves of N-doped rGO at 1 mA. (e, f) GCD curves of N-doped rGO at different currents.

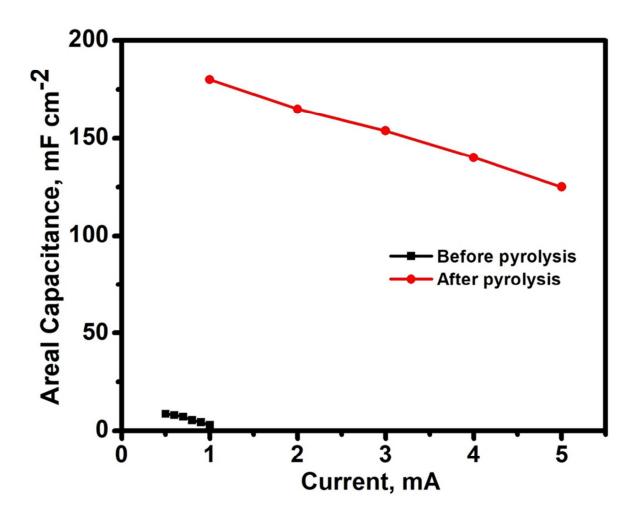


Fig. S8 The calculated areal capacitance of N-doped rGO at different currents.

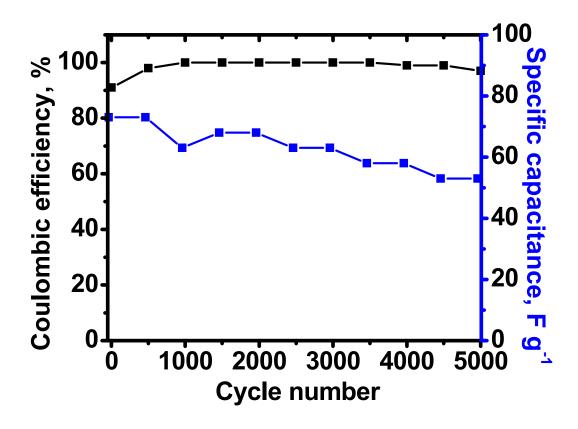


Fig. S9 The cycling life of the hybrid supercapacitor over 5000 cycles.