

Supporting Information

Metastability-induced high pseudocapacitance for FeCN₂@NC anode materials as sodium ion batteries

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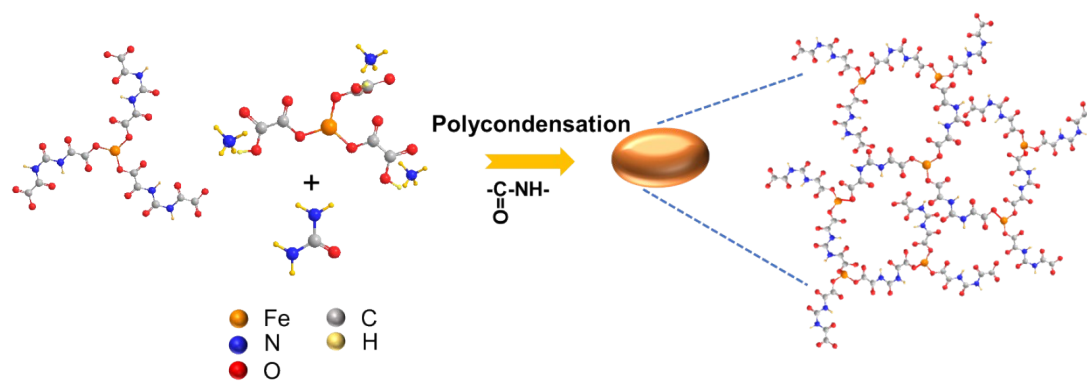
Scheme S1. Detailed illustration of the formation of net-like gel

Figure S1 (a) XRD patterns of FeCN₂@NC after half a year of exposure to air
(b) Raman spectrum
(c) XPS survey spectra of FeCN₂@NC;
(d) XRD patterns of NC substrates
(e) element mapping nitrogen doped carbon (NC) substrates
(f) photo graph of the gel obtained at 200 °C
(g) XRD patterns of the intermediate products obtained at 200 and 400 °C
(h) FTIR spectra of the intermediate products obtained at 200 and 400 °C

Figure S2 TGA curve of FeCN₂@NC composites

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Figure S4 Comparison of iron-based sodium-ion anodes in this work and previous reports



Scheme S1. Detailed illustration of the formation of net-like gel

The polyamidation reaction between of -COOH of ammonium ferric oxalate and -NH_2 of urea takes place to form gel composed of net-like iron-complex molecular framework during heating under Ar atmosphere.

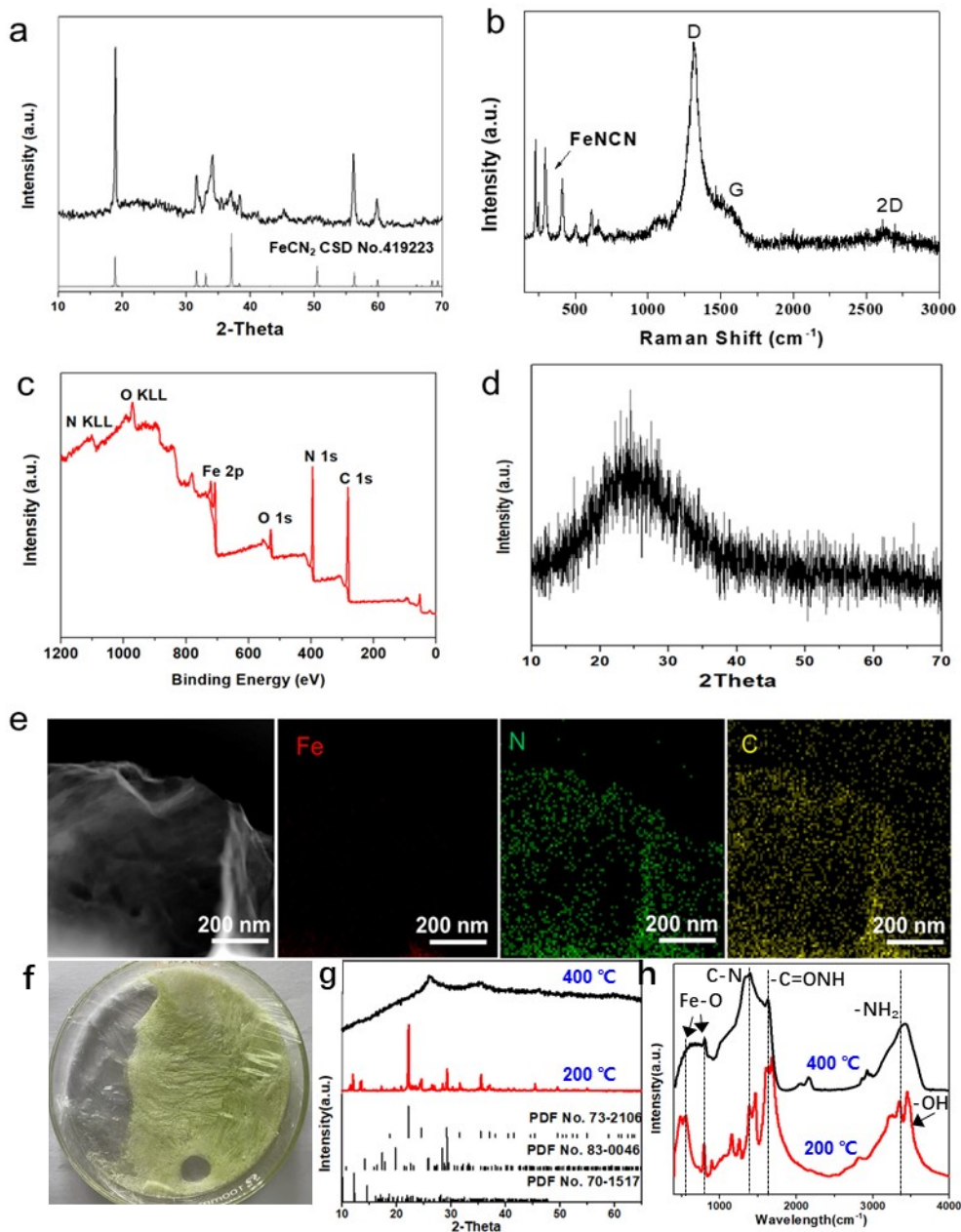


Figure S1 (a) XRD patterns of $\text{FeCN}_2@\text{NC}$ after half a year of exposure to air (b) Raman spectrum (c) XPS survey spectra of $\text{FeCN}_2@\text{NC}$; (d) XRD patterns of NC substrates (e) element mapping of Fe, N and C for nitrogen doped carbon (NC) substrates (f) photo graph of the gel obtained at 200 °C (g) XRD patterns and (h) FTIR spectra of the intermediate products obtained at 200 and 400 °C

The XRD patterns of $\text{FeCN}_2@\text{NC}$ (Figure S1a) which is exposed to air for half a year

can be indexed to FeCN_2 , indicating there is no obvious phase transformation, confirming improved phase stability after carbon coating. Raman spectra (Figure S1b) and the XRD patterns of the substrates show the carbon in the composites are mainly amorphous. The element mapping results reveal N and C elements are contained in the substrates. Figure S1(f) shows the gel obtained at 200 °C. The XRD patterns shows intermediate products obtained at 200 °C mainly includes urea (PDF No. 73-2106), compounds of C, N and O (PDF No. 83-0046) and complex of iron and C,N and O(PDF No.70-1517). When the temperature raised to 400 °C, the mixture transformed to amorphous carbon (Figure S1(g)). Figure S1(h) shows the FTIR spectra of the intermediate products. There are plenty of $-\text{NH}_2$, $-\text{OH}$ organic groups ($3200\text{-}3500\text{ cm}^{-1}$) in the compounds. The strong adsorption around 1700 cm^{-1} could confirm the existence of $\text{C}=\text{O}-\text{NH}-$ group in the iron-complex. The peak at 1375 cm^{-1} could attribute to the C-N bond in the compounds. The peaks ranging from $500\text{-}790\text{ cm}^{-1}$ are related to Fe-O bonds in the Fe-complex.

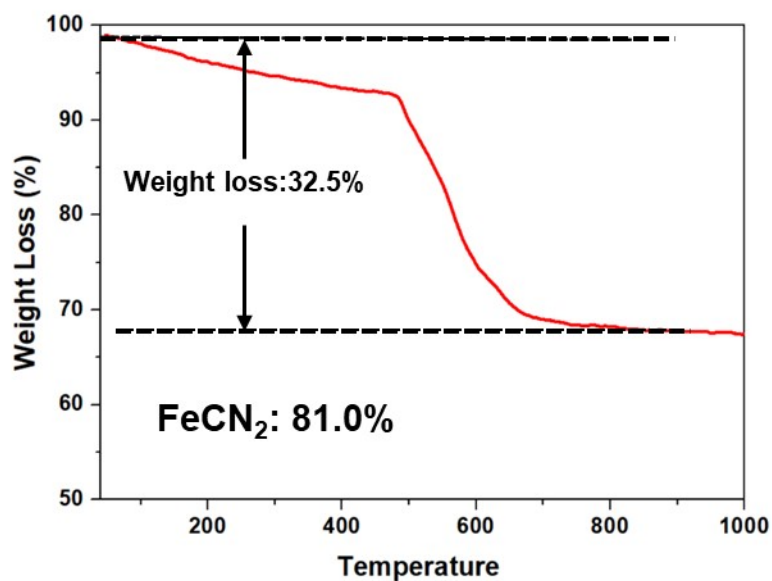


Figure S2 TGA curve of FeCN₂@NC composites

TGA analysis is carried out in air to determine the proportion of FeCN₂ in the composites. According to the remaining mass of oxidized products (Fe₂O₃), the FeCN₂ content is calculated about 81% .

$$\text{Mass (Fe element)} = \text{Mass(Fe}_2\text{O}_3) * 0.7 = \text{Mass((FeCN}_2\text{)} * 0.583$$

$$\text{Mass (FeCN}_2\text{)} = \text{Mass((Fe}_2\text{O}_3) * 0.7 / 0.583 = 0.81$$

Consequently, FeCN₂ accounts for 81% in FeCN₂@NC composites.

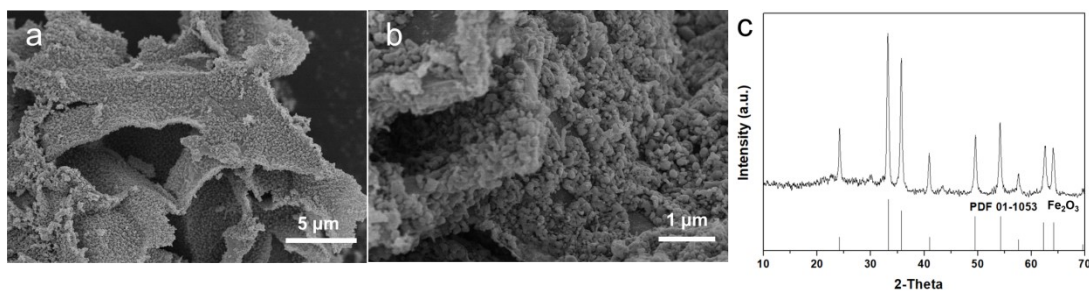


Figure S3 (a, b) SEM images and (c) XRD patterns of reference sample $\text{Fe}_2\text{O}_3@\text{NC}$

The reference sample $\text{Fe}_2\text{O}_3@\text{NC}$ is obtained by the oxidation of $\text{FeCN}_2@\text{NC}$ in air heating at 300 °C for 2 hours at a heating rate of 5 °C/min. Figure S2(a, b) show a morphology of NC substrate supported Fe_2O_3 structure, which is similar to $\text{FeCN}_2@\text{NC}$.

The XRD patterns of the composites is fully consistent with the standard iron oxides (PDF # 01-1053).

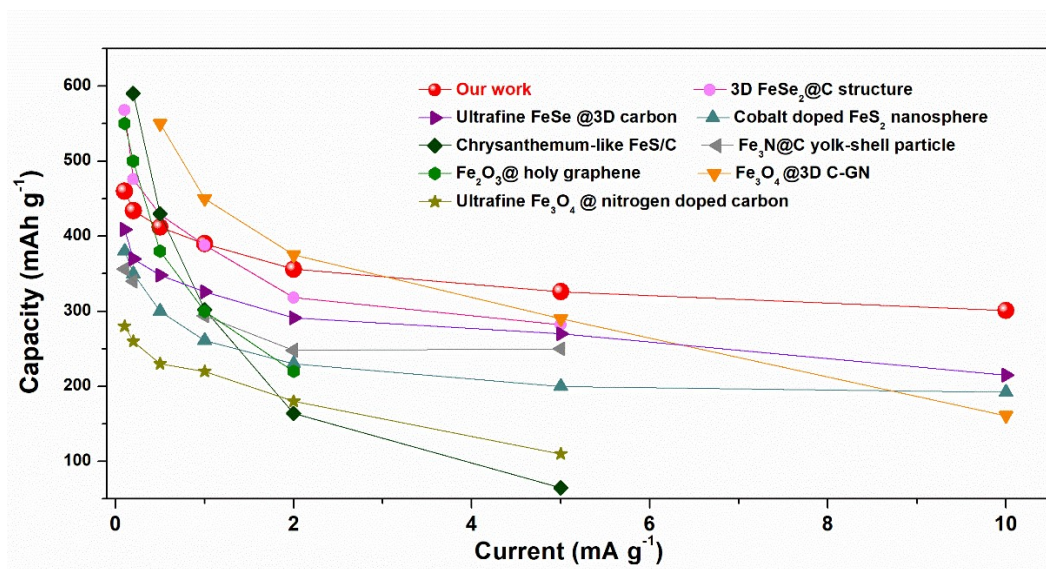


Figure S4 Comparison of iron-based sodium-ion anodes in this work and previous reports^[1-8]

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