FeSe\textsubscript{2} nanocrystalline aggregated microsphere with ultrahigh pseudocapacitive contribution for enhanced sodium-ion storage

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Material characterization

The morphologies and microstructure of as-synthesized samples were observed by using field emission scanning electron microscopy (FESEM, JSM-7001F, 10 kV), transmission electron microscopy (TEM, JEM-2100 operated at 200 kV) and energy dispersive spectroscopy (EDS, JEM-ARM200F). Crystalline structures of materials were characterized in the range of 10-80° (2θ) through German Bruker D8 Advance X-ray diffractometer (XRD) using Cu Kα (λ=1.54056 Å) radiation. The chemical compositions and elemental valence of synthesized samples were investigated by the X-ray photoelectron spectrometer (XPS, Thermo Fisher Scientific, USA).

Electrochemical measurements

The electrochemical properties of the obtained FeSe2@NC materials were tested by assembling CR2032 coin cells in an Ar–filled glovebox using FeSe2@NC as the working electrode, sodium metal as the counter electrode, 1.0 M NaClO4 dissolved in a mixture of EC, DMC and EMC (1:1:1, vol.%) with 2 vol.% FEC as electrolyte, and Whatman glass fiber as separator. The working electrodes were fabricated as follows: first, the obtained FeSe2@NC materials were mixed with Ketjen black (as conductive agent), and poly (vinylidene fluoride) (PVDF, as binder) in a mass ratio of 7:2:1, the mixture was dispersed in N-methyl-2-pyrrolidone (NMP) to form homogeneous slurry, and then the resulting slurry was uniformly coated on aluminum foil and dried at 120 °C for 12 h in a vacuum oven. Finally, the FeSe2@NC coated Al foils were punched into 14 mm disks to obtain the working electrodes. The loading mass of active material was about 1 mg. The electrochemical measurements, including constant current charge-discharge testing, cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were performed using Land battery testing system (CT4008TNn-5V10mA-164, China) and an electrochemical workstation (CHI660E, China), respectively. The cells were galvanostatically charged and discharged between 0.5 and 3.0 V at various current densities. The capacity was calculated according to the mass of the active material in the electrode, excluding the mass of conductive carbon and binder.
Fig S1. XRD patterns of FeSe$_2$-0, FeSe$_2$@NC-0.1, FeSe$_2$@NC-0.2, FeSe$_2$@NC-0.3 and FeSe$_2$@NC-0.5

Fig. S2 TEM images of FeSe$_2$@NC-0.3

Fig S3. TEM images of FeSe$_2$-0
**Fig S4.** Rate capability of FeSe$_2$-0

**Fig. S5** Relation between log $i_{\text{peak current}}$ and log $v_{\text{scan rate}}$ of FeSe$_2$@NC-0.3
Fig. S6 The fitting diagrams of the normalized contribution rate of capacitance at different sweep rates for FeSe$_2$@NS-0.3
Fig. S7 CV curves of FeSez-0 at various scan rates (a) and normalized contribution ratio of capacitive at different scan rates (b)
Fig. S8 The fitting diagrams of the normalized contribution rate of capacitance at different sweep rates for FeSe$_2$-0
Fig. S9 Nyquist plots of FeSe$_2$-0 and FeSe$_2$@NC-0.3 electrodes