Supplementary Information

1. Experimental sections

1.1. Materials and synthesis

The chemical Ammonium metavanadate, sodium dodecyl benzenesulfonate (SDBS), Ammonium persulfate, potassium hydroxide, sodium hydroxide, N-methl-2-pyrrolidone (99.9%), Anhydrous ethanol (99.7%), Methanol (99.7%), Activated carbon (AC), Biotetrafluoroethylene and thioacetamide (TAA) are from purchased ChengDu Chron Chemicals Co,.Ltd (China) Biochemical Diatomite (De) and pyrrole (98%) were purchased from Tianjin Damao and Shanghai Kefeng chemical reagent factory in China, respectively. All the reagents used were of analytical reagent grade and no purification was carried out.

1.1.1. Pretreatment of diatomite

The surface of the diatomite must be pretreated to increase its activity. In general, weigh 4 g of diatomite into 100 mL of 1 M sodium hydroxide and stir at 60 °C for one hour. After this period, the solution must be thoroughly washed completely and the diatomite centrifuged three separate occasions with dehydrated alcohol. Subsequently, material should be baked in an oven at a temperature of 110 °C.

1.1.2. Preparations of De@VS_4

De@VS_4 was synthesized using a streamlined one-step hydrothermal method. In the typical procedure, 50 mg of pretreated diatomite was accurately weighed and dispersed into 70 ml of methanol, then vigorously stirred for one hour to ensure uniform distribution in the solution. Then, 0.44 g of ammonium metavanadate and 1.42 g of thioacetamide were added to the solution. The prepared mixture was subsequently placed into a 100 mL Teflon-lined stainless-steel autoclave for further processing. Following the thermal treatment, the autoclave was left to cool naturally to room temperature. The resultant precipitate was subjected to washing and centrifugation with ethanol and water in five consecutive cycles, followed by drying at 60 ° C for a duration of four hours to yield the final De@VS_4.

1.1.3. Preparation of De@VS_4@PPy

De@VS$_4$@PPy was obtained by one-step oxidative polymerisation of De@VS$_4$. Briefly,1 g of sodium dodecyl benzenesulfonate and 100 mg of De@VS$_4$ are measured and combined with 50 mL of deionized water. The mixture is then stirred for a duration of 30 minutes to facilitate complete dissolution and the formation of a suspension. Then 125 μL pyrrole was added to the suspension and the ice bath was used for 1 hours. The suspension generated in the preceding step was subsequently combined with 5 mL of a 0.1 mol/L ammonium persulfate solution and agitated in an ice bath for a duration of 12 hours to yield De@VS$_4$@PPy. The resultant product was subjected to multiple rinsed with deionized water and alcohol, then dried in an oven at 60 °C for 5 hours to prepare it for further testing. De@PPy was synthesized through a one-step oxidative polymerization process directly on pretreated diatomite.

1.2. Material characterization

The X-ray diffraction (XRD) patterns of the samples were characterized by using an X-ray powder spectrophotometer (Rigaku Ultima IV, Rigaku Corporation, Japan) using Cu Kα radiation at λ =1.5406 Å. The X-ray photoelectron spectroscopy (XPS) spectra were performed on a Thermo Fisher Scientific K-ALPHA instrument with an excitation source of Al Kα. The Raman spectra of the samples were precisely characterized using a Thermo Fisher DXR3 spectrometer with 532 nm excitation. The microstructural and morphological characteristics of the samples were meticulously examined using advanced focused ion beam scanning electron microscopy (FIB/SEM, ZEISS AURIGA) and high-resolution transmission electron microscopy (TEM, Talos F200S).

1.3. Electrochemical characterizations

The analysis of the prepared samples with three-electrodes and two-electrodes was carried out by means of an electrochemical workstation (CHI 660E). The electrolyte used was 1 M potassium hydroxide.

Three-electrodes configuration were fabricated using the samples as working electrodes, with a platinum (Pt) electrode serving as the counter electrode and a standard calomel electrode (SCE) as the reference electrode. For the working electrodes, the prepared samples, AC, and PVDF were initially dissolved in NMP, forming a homogeneous slurry with a precise mass ratio of 7:2:1. Then the slurry is evenly painted onto a 1x1 cm^2 foam nickel. Finally, the slurry-coated nickel foam was placed in an oven at 120 °C for 12 h to facilitate solvent evaporation. The mass loading of the active material on the working electrode is approximately 2 mg/cm^2 . The specific capacitance, rate capability, and cycling stability of the fabricated working electrodes were evaluated using cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) measurements. The internal resistance of the working electrode was evaluated through electrochemical impedance spectroscopy (EIS) conducted on the three electrodes over a frequency range of 0.01 Hz to 100 kHz.

In the two-electrode configuration, an asymmetric supercapacitor was assembled using Whatman filter paper as the separator, activated carbon as the negative electrode, and De@VS_4@PPy as the positive electrode. The slurry was applied to the nickel foam following the same procedure used for the three-electrodes. The galvanostatic charge-discharge measurement has an expanding potential window of 0-1.8 V.

The specific capacitance (C_m, F/g) of the electrodes was calculated using galvanostatic charge-discharge measurements, and the corresponding formula is provided below:

$$Cm = \frac{I \not{\beta} \triangle t}{m \times \triangle V} \tag{1}$$

where I is the discharging current (A), $\triangle t$ is the discharge time (s), $\triangle V$ is the potential window (V), m is the weight of active material (g).

The specific capacitance for the two-electrode is calculated using the same formula as that for the three-electrode configuration. The power density (P, W/kg) and energy density (E, Wh/kg) of the two-electrodes are derived using the following equations:

$$E = 1/2 \times Cm \times V^2/3.6 \tag{2}$$

$$P = 3600 \times E/\triangle t \tag{3}$$

where m represents the total mass of the two electrodes (g) and V is the potential window (V). $\triangle t$ is the discharge time (s).

Figure S1.SEM image of (a) De@PPy, (b) De@VS_4 and (c) De@VS_4@PPy.

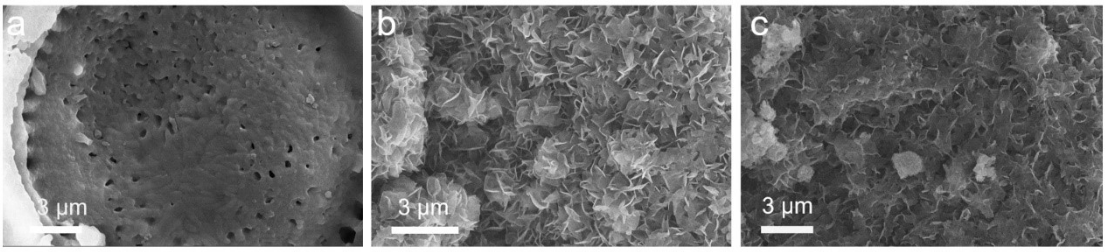

Figure S2. EDS mapping of De@VS$_4$@PPy.

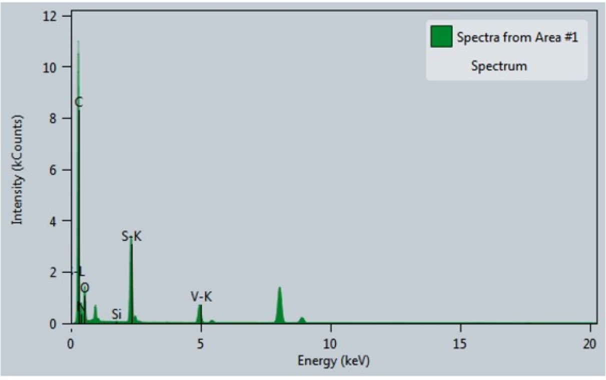

Table 1
EDS mass content.

Element	Weight percentage (%)	Atomic percentage (%)
C	62.83	79.5
N	2.20	2.38
O	6.00	5.7
Si	0.15	0.08
S	21.31	10.10
V	7.52	2.24

Figure S3. Total survey spectrum and high-resolution (b) C 1s, (c) Si 2p and (d) O 1s XPS spectra of De@VS$_4$@PPy.

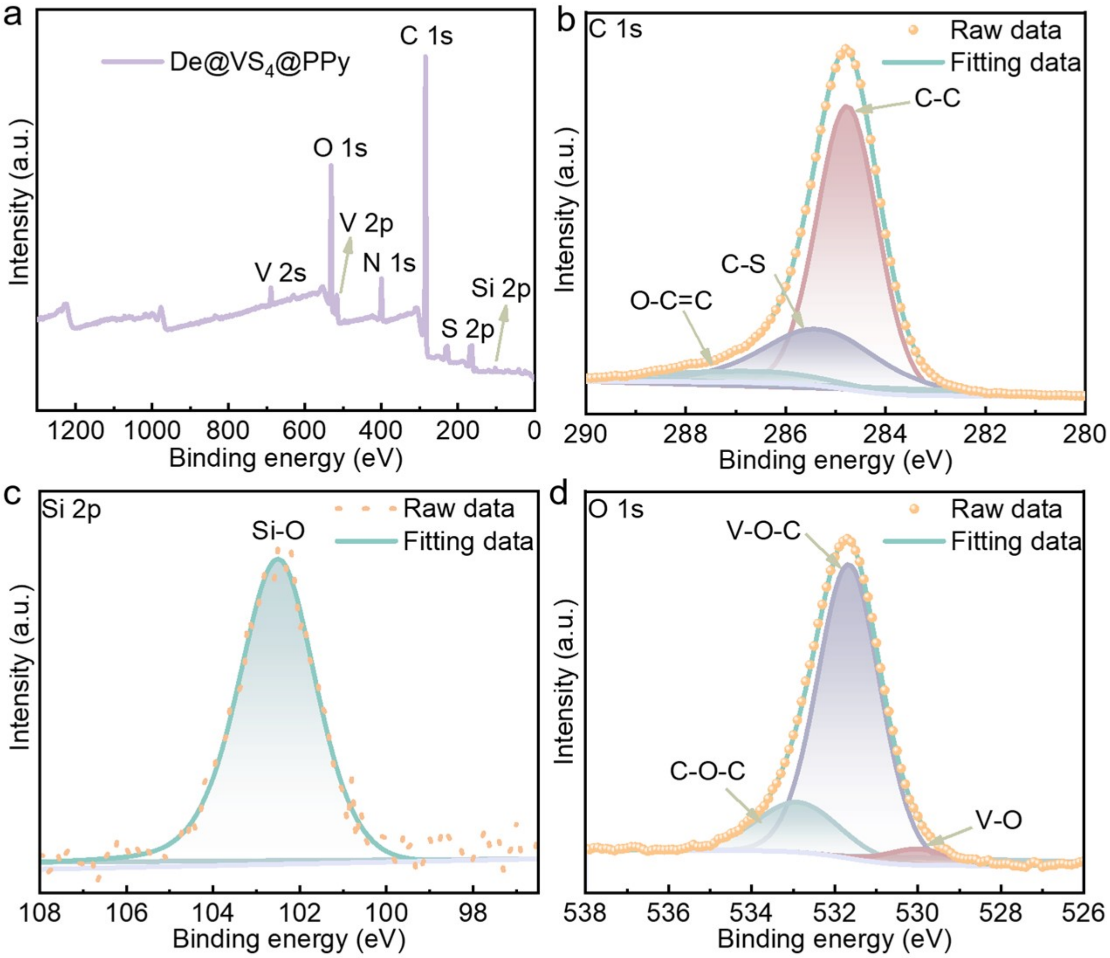

Fig. S4 The (a) CV curves at different scan rates and (b) CC curves at different current density of De@VS_4@PPy.

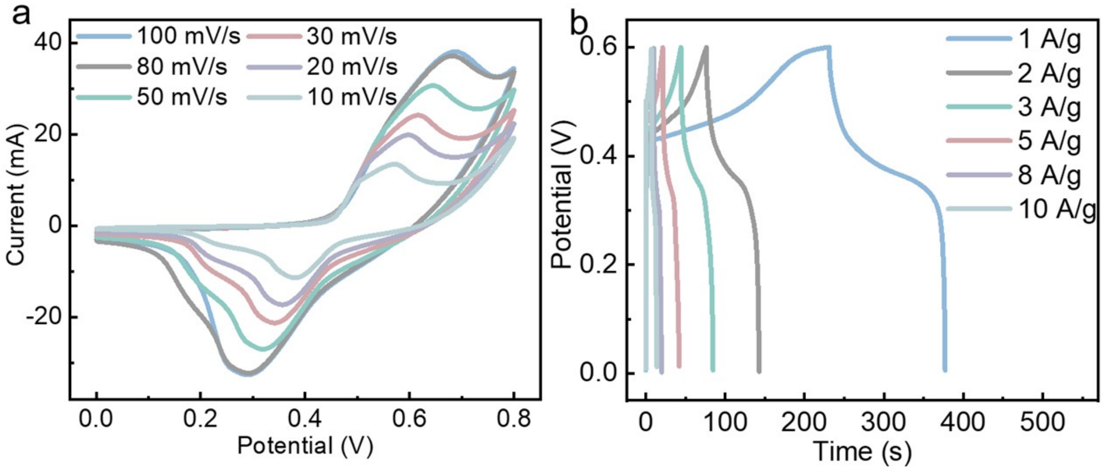

Fig. S5 The (a) CV curves at different scan rates and (b) CC curves at different current density of De@PPy.

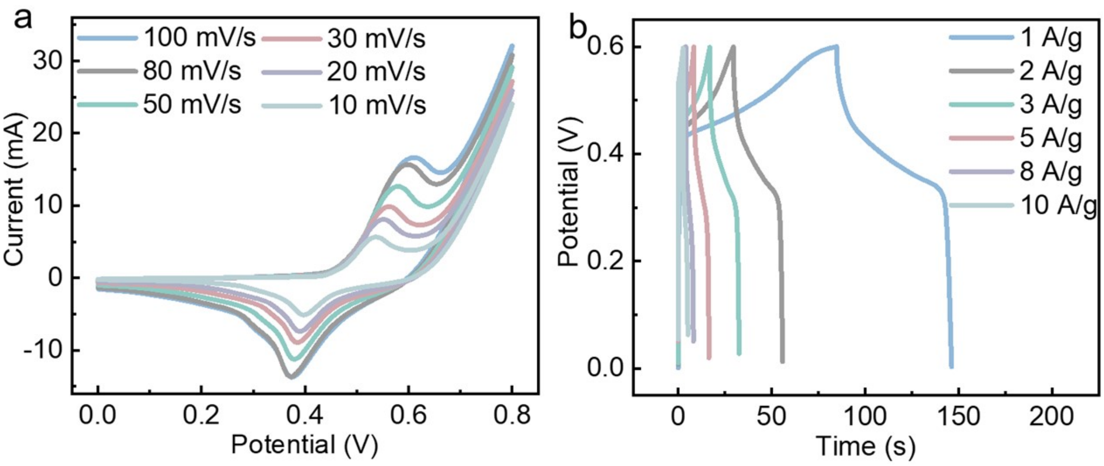

Fig. S6 The (a) CV curves at different scan rates and (b) CC curves at different current density of De@VS_4.

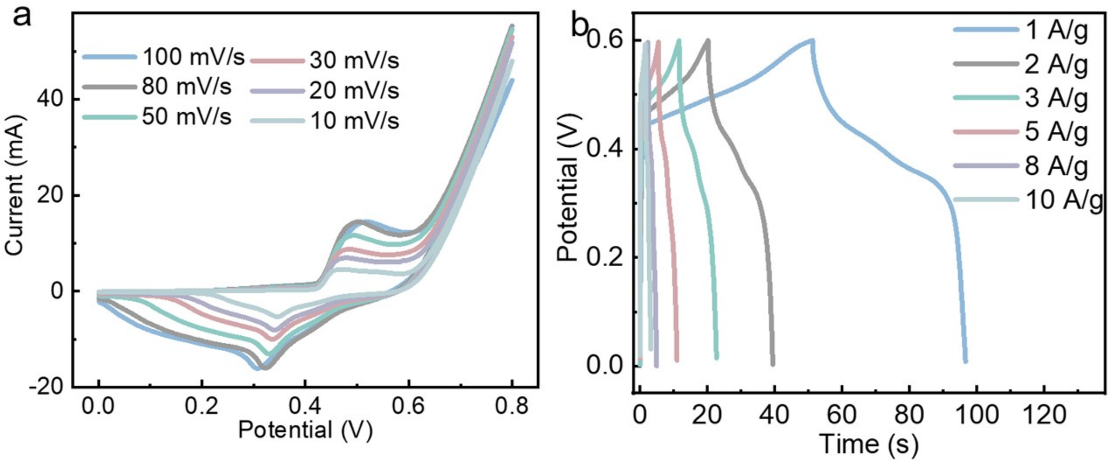

Fig. S7 The comparison of the original and fitted CV curves for De@VS_4@PPy at scan rates of 1,2,3,4 and 5 mV/s, respectively.

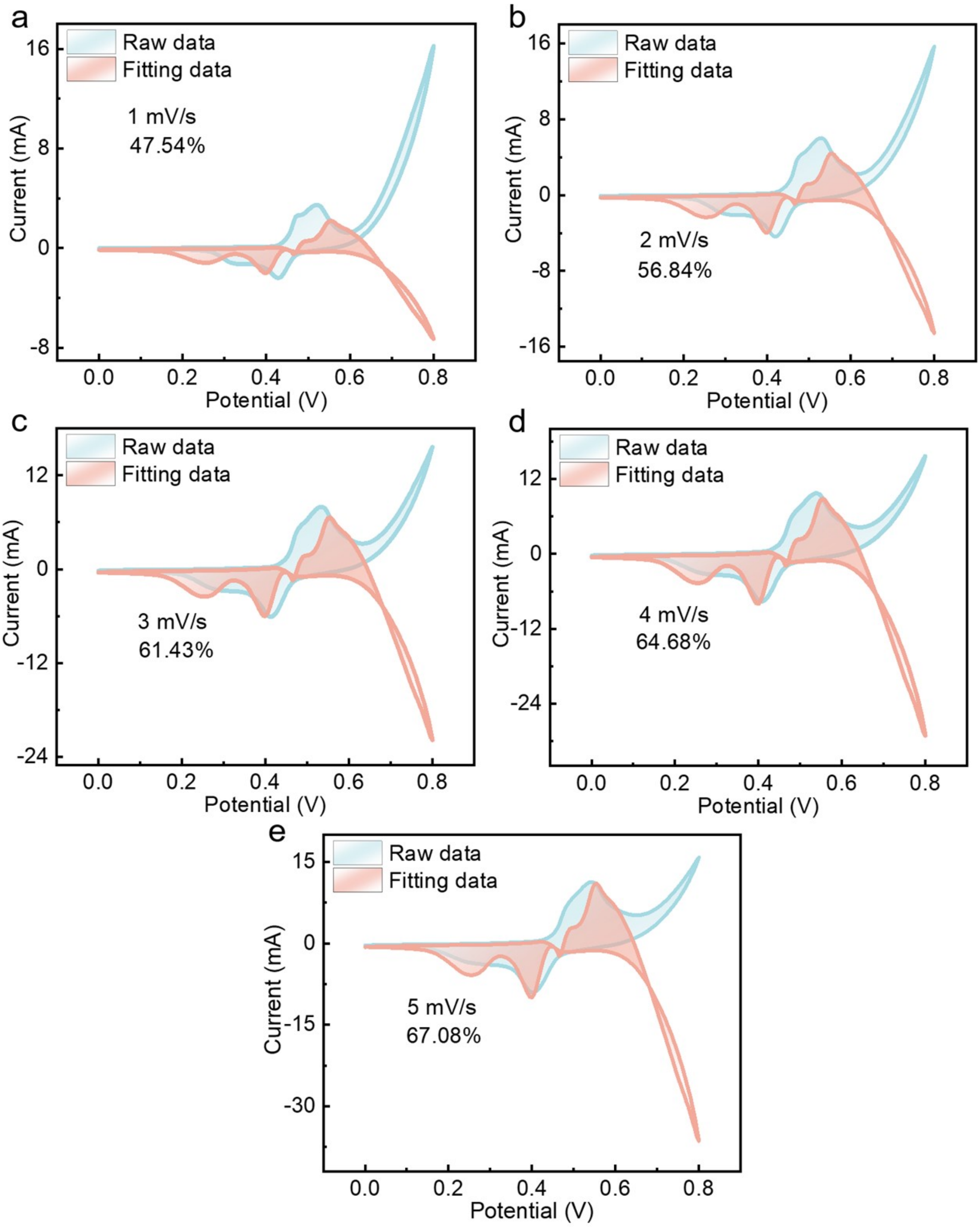

Figure S8 Enlarged view of the Nyquist plot of the before and after cycling of De@VS$_4$@PPy//AC device.

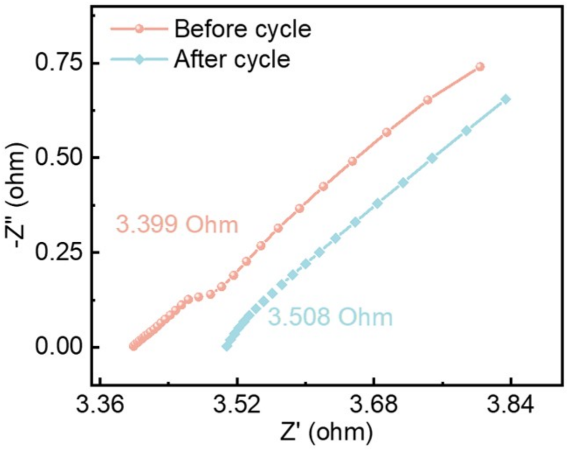

Figure S9 The CV curves of AC and De@VS_4@PPy at 100 mV/s in a three-electrode system. (b) Rate capability of De@VS_4@PPy//AC at different current densities.