

Porous diatomite-mixed 1,4,5,8-NTCDA nanowires as high-performance electrode materials for lithium-ion batteries

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1. Solubility analysis of NTCDA active substances in electrolyte solvents

In order to investigate the solubility of NTCDA active substances in electrolyte solvents, we designed a mixed liquid of NTCDA and EC/DMC which can theoretically reach the concentration of 1×10^{-5} M. A certain weight of 1,4,5,8-NTCDA was added to a certain volume of EC/DMC electrolyte solvent, and after 1 h of storage, their UV-Vis absorption spectra were measured by two different types of UV-Vis spectrophotometer, as shown in **Fig. S1 a-b**. It can be seen that for the pristine 1,4,5,8-NTCDA powder, after 1 h of storage, there is a small amount of dissolution ($\lambda_{\max}=358$ nm) in the electrolyte of EC/DMC, and the absorbance can reach about 0.1, indicating that 1,4,5,8-NTCDA has a small solubility in the electrolyte of EC/DMC solvent. However, for the three different electrodes, although in theory, the weight of the active substance NTCDA in their electrodes is enough to form the concentration of 1×10^{-5} M, their solubility is very small. After soaking for an hour, only a small amount of dissolution and absorbance do not exceed 0.02, indicating that the active substances in the electrode have better stability.

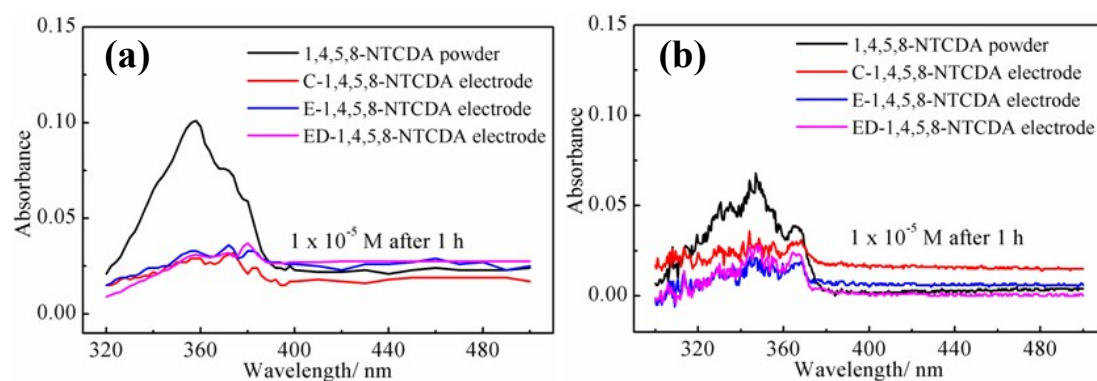


Fig. S1 UV-Vis absorption spectra of 1,4,5,8-NTCDA powder and three different NTCDA electrodes soaking in electrolyte solvents (theoretically up to 1×10^{-5} M). (a) tested by 722 type UV-Vis spectrophotometer; (b) tested by AC1410002 manual UV-Vis spectrophotometer.

With regard to inhibiting the dissolution of small molecular organic compounds in electrolyte, we have carefully read some literature (*Chem*, 2018, 4, 2600; *Angew. Chem. Int. Ed.*, 2018, 57, 16072; *Adv. Mater.*, 2016, 28, 9182) and obtained a lot of inspiration. Among them, parasitic organic small molecules on inorganic carriers is an effective way to reduce solubility. In this way, after the 1,4,5,8-NTCDA is mixed with the S-P conductive agent and the PVDF binder, the slurry is made and electrospun onto the copper foil, and the solubility of the slurry will be greatly suppressed under the fixation of the S-P and the PVDF. This conclusion is confirmed by our experimental results, as shown in **Fig. S1**. It can be seen that the NTCDA active substances in the three electrodes are basically not dissolved in the electrolyte solvent, and the absorption peak can not be seen at the 358 nm. The results show that the solubility of NTCDA active substances can be effectively suppressed under the action of conductive agent and binder after the electrode is prepared.

2. Morphology, composition and capacity contribution of diatomite

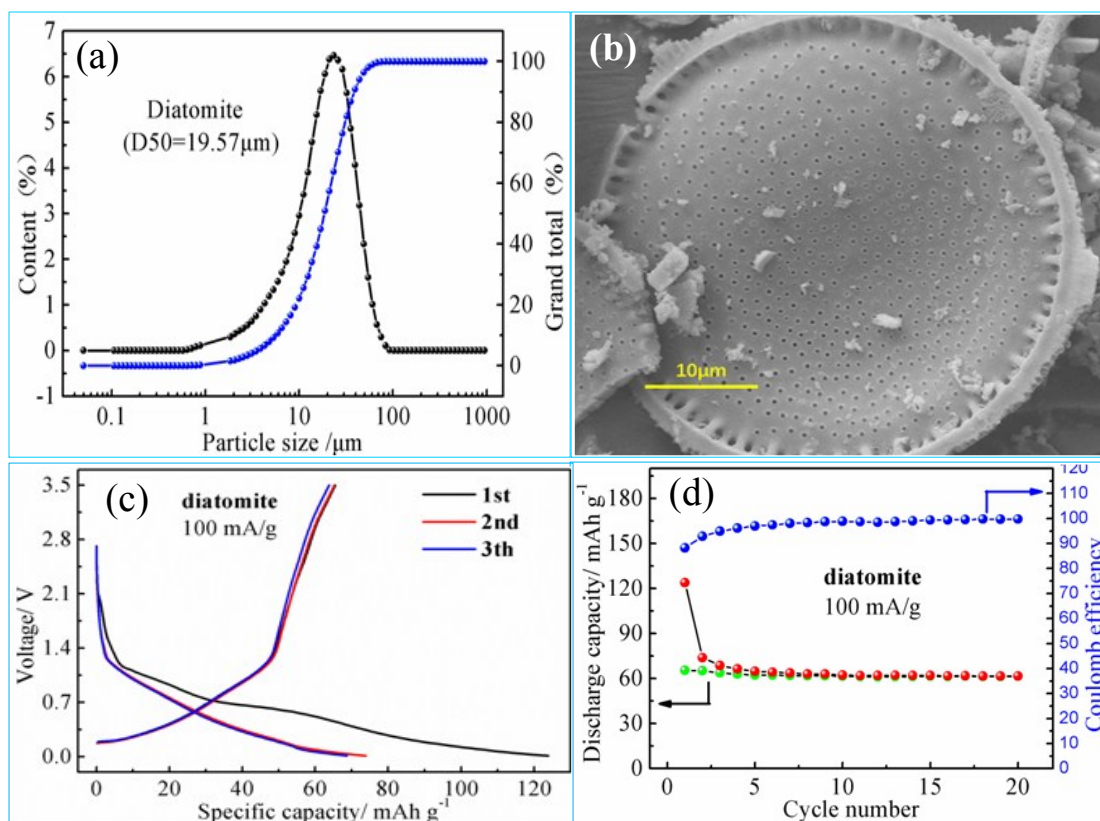


Fig. S2 (a) Particle size distribution curve of diatomite; (b) SEM image of diatomite; (c) The second charge-discharge curve of diatomite; (d) Charge-discharge cycle performance of diatomite.

The content of diatomite in the composite is 40%. It is verified that the diatomite used in this manuscript was purified, and Elemental Analysis shows that it is composed of 98.05% of SiO_2 , 0.32% of Fe_2O_3 and 0.52% of K_2O and Na_2O , 0.12% of CaO , 0.05% of TiO_2 , 0.02% of MgO and 0.92% of other ingredients (**Table S1**).

Table S1 Ingredients of diatomite after purified

Ingredients/%	SiO_2	MgO	Fe_2O_3	K_2O	Na_2O	CaO	TiO_2	Other
After purified	98.05	0.02	0.32	0.26	0.26	0.12	0.05	0.92

It can be seen from **Fig. S2 a-b** that the diatomite used in our manuscript has a large size of about 20 μm . In order to find out the contribution of diatomite to the capacity, we mixed diatomite (70%) with conductive agent super P (20%) and PVDF binder (10%) to make slurry, and coated on the surface of copper foil. Then the lithium sheet is used as the counter electrode, and half batteries are assembled to investigate their charge/discharge performance, as shown in **Fig. S2 c-d**. It is found that the capacity of diatomite is only 60-70 mAh/g , which contributes little to the whole negative capacity.

3. Contrast of SEM images before and after a certain number of cycles

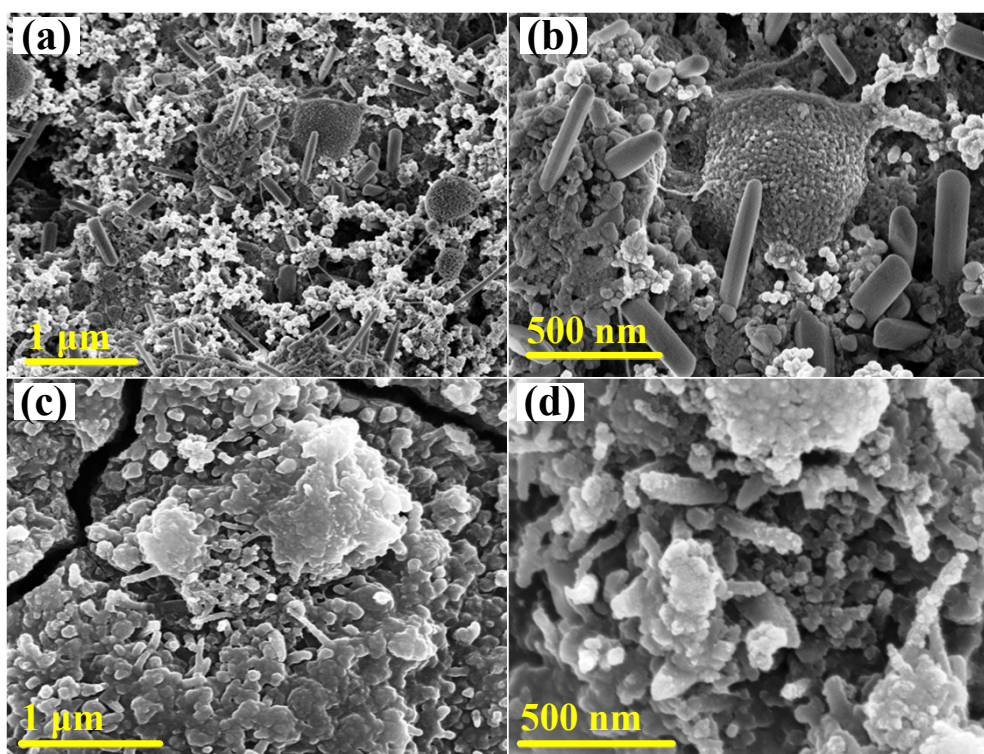


Fig. S3-2 SEM images of electrospinning nanorods electrode (C-1,4,5,8-NTCDA) before and after 200 cycles

It can be seen from Fig. S3-1 that many NTCDA nanoblocks (500-800 nm) are distributed in the newly prepared C-1,4,5,8-NTCDA electrodes (Fig. S3-1 a-b). However, after 400 cycles, the active particles of NTCDA have obvious powder marks, and a lot of cracks and pore structures are formed on the surface, and even some of the active particles crack (Fig. S3-1 c-d). This process will inevitably expose more active particles and increase the active point position of the electrode material. Similarly, for the E-1,4,5,8-NTCDA electrode, it can be seen that many NTCDA nanorods (50-100 nm) are distributed in the newly prepared electrodes (Fig. S3-2 a-b). After 200 cycles, there is also a significant decrease in the number and size of NTCDA active particles in the shape of nanorods (Fig. S3-2 c-d). With the increase of cycle, the diameter and length of NTCDA nanorods tend to decrease obviously. Therefore, for large particles of organic materials, in the process of cycling, lithium ion is constantly embedded and removed from the electrode material, the material particles will gradually become smaller particles, the specific surface area of the active material increases, and the active point site increases, which will inevitably lead to the gradual increase of capacity.

In general, with the progress of the cycle, there are two main reasons for the gradual increase of the capacity of the electrode. On the one hand, as the circulation is carried out, the NTCDA nanorod is continuously refined and pulverized, and more active points are exposed; on the other hand, more pore structures are formed in the electrode material, and the active point is further increased, and the transmission channel of the lithium ion is increased. In this case, the capacity of the battery is gradually increased to be necessary.

4. EIS Analysis of three different electrodes

In order to further verify the impedance value of electrodes with different preparation technology, the impedance of another batch of three batteries was measured on another type of electrochemical workstation, as shown in Fig.S4 and Table S2.

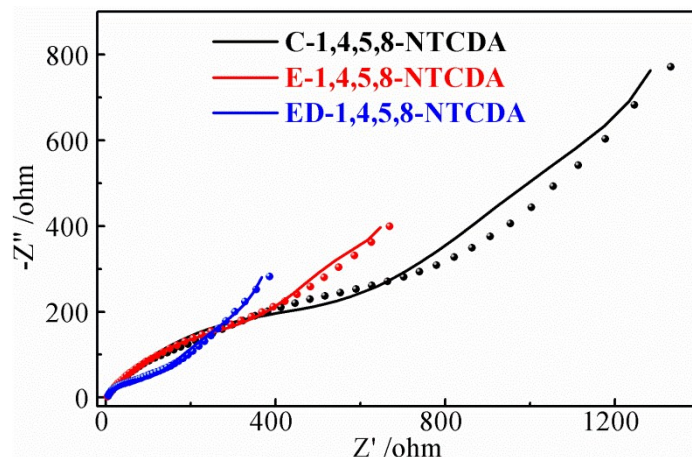


Fig. S4 EIS spectra of NDCTA electrodes formed using different preparation technologies

Table S2. List of EIS fitting parameters of the three electrodes with different preparation technology

Electrodes	R_1/Ω (error)	$R_2(R_{CT})/\Omega$ (error)	W_1-R/Ω (error)	W_1-T/s (error)	W_1-P (error)
C-1,4,5,8-NTCDA	1.65 (15.6%)	511.3 (14.1%)	2619 (11.8%)	8.49 (29.9%)	0.48 (8.9%)
E-1,4,5,8-NTCDA	2.72 (2.70%)	321.2 (10.3%)	1314 (7.2%)	0.96 (16.1%)	0.56 (6.1%)
ED-1,4,5,8-NTCDA	2.37 (18.5%)	65.50 (11.4%)	855.9 (7.6%)	0.30 (17.7%)	0.45 (4.2%)

It can be seen that the charge transfer impedance (R_2) and diffusion impedance (W_1-R) tested by this type of electrochemical workstation still show the same rules as the impedance value discussed above, that is, the **ED-1,4,5,8-NTCDA** electrode has the smallest charge transfer impedance (R_2) and diffusion impedance (W_1-R). Moreover, the W_1-T ($W_1-T=L^2/D$, Where L is the effective diffusion layer thickness and D is the one-dimensional diffusion coefficient) represents the Warberg coefficient has also investigated. It is clear that the **ED-1,4,5,8-NTCDA** electrode mixed with diatomite has the smallest W_1-T value, indicating that the **ED-1,4,5,8-NTCDA** electrode has the maximum one-dimensional diffusion coefficient of lithium ion, which is of great help to the performance of the battery. The EIS impedance fitting data of different electrode batteries show that the composite electrode materials have faster lithium ion transport channels.

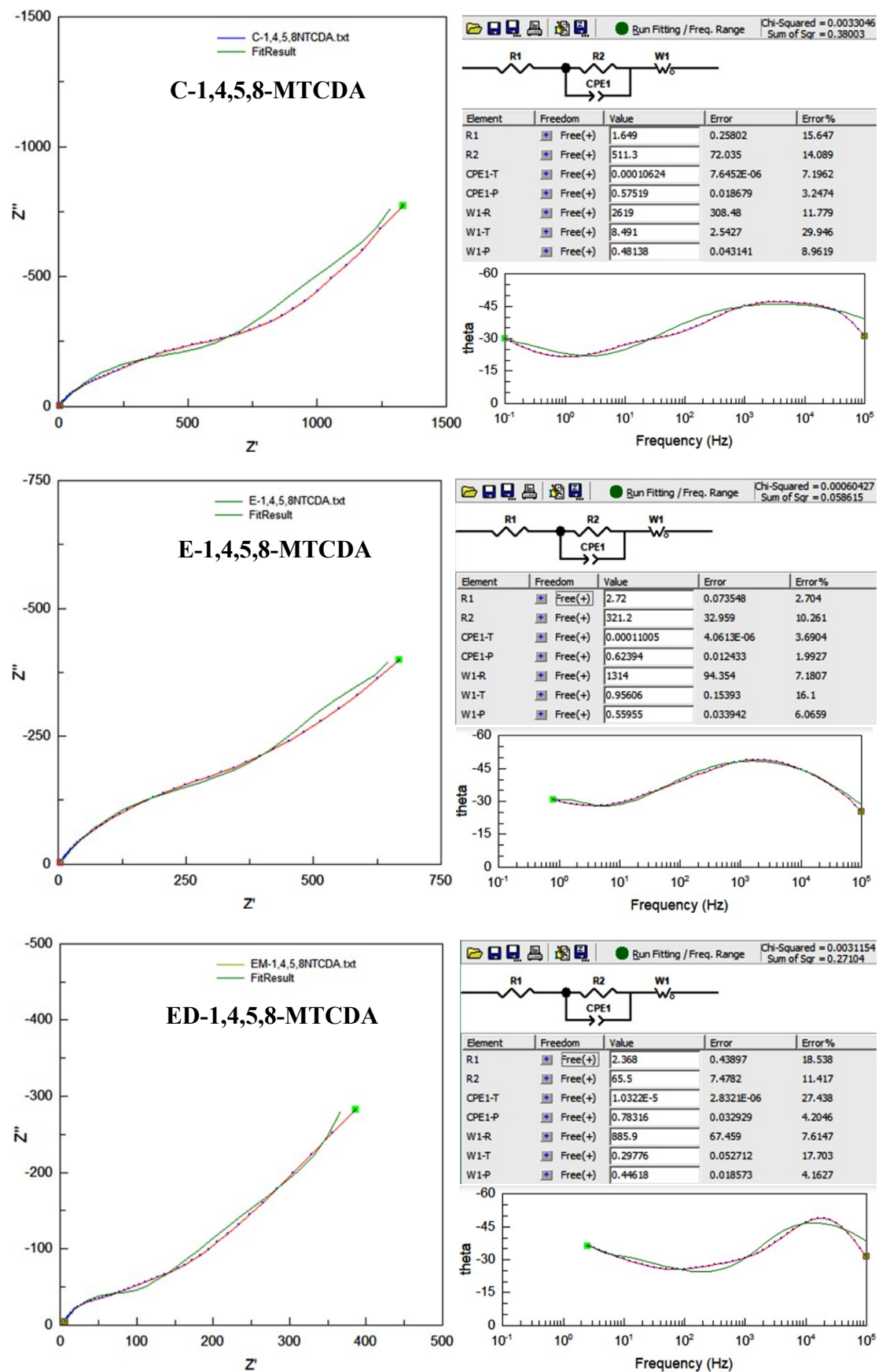


Fig. S5 EIS fitting Circuit and fitting Curves of NDCTA electrodes prepared using different preparation technologies