Toward High Energy-density and Long Cycling-lifespan Lithium Ion Capacitor: A 3D Carbon Modified Low-potential Li_2TiSiO_5 Anode Coupled with A Lignin-derived Activated Carbon Cathode

Liming Jin, Ruiqi Gong, Weichao Zhang, Yue Xiang, Junsheng Zheng [*], Zhonghua Xiang[*], Cunman Zhang, Yongyao Xia, and Jim P. Zheng[*]

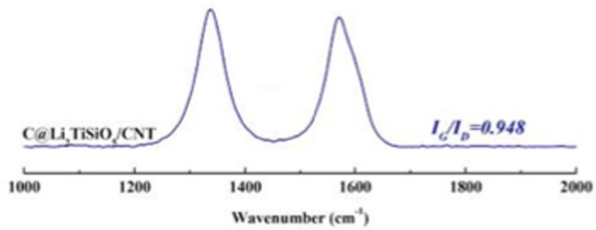

Figure S1. The Raman spectrum of 3DC@LTSO.

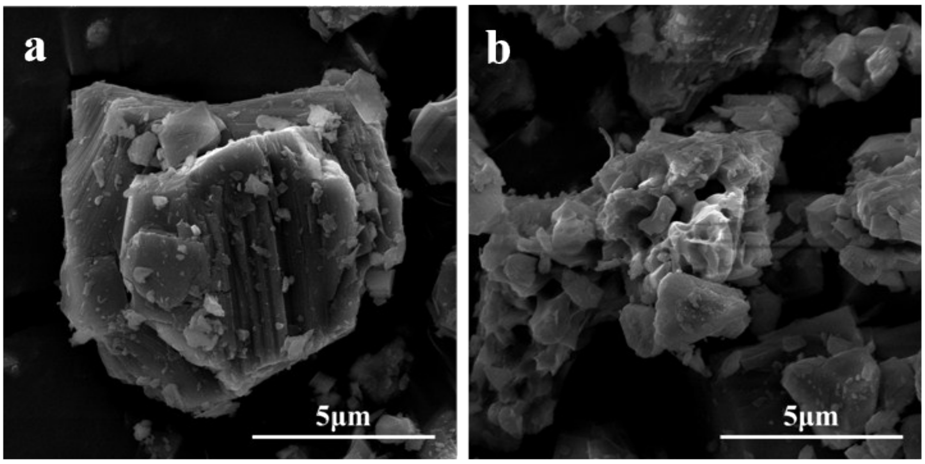

Figure S2. FESEM microscopy of pure LTSO samples.

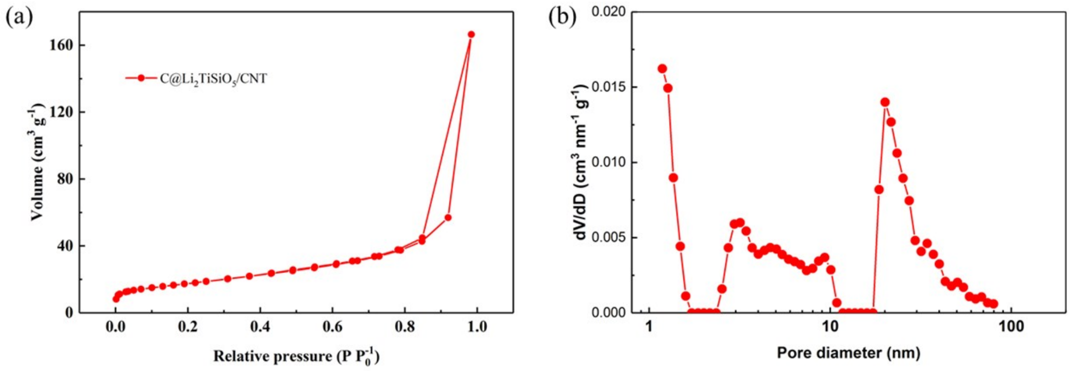

Figure S3. (a) Nitrogen adsorption-desorption isotherms and (b) the corresponding pore size distribution of 3DC@LTSO composite.

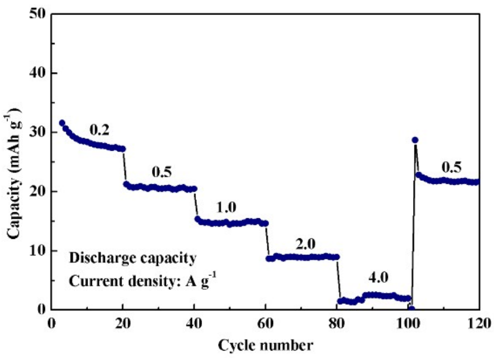

Figure S4. Multi-rate performance of pure LTSO electrode.

S5. Li+ diffusion co-efficient during charge and discharge

Randles-Sevcik equation, which is always used to calculate the Li+ diffusion coefficient during charge and discharge, is represented in Equation:

$$I_p = 2.69 \times 10^5 \, n^{3/2} \, A \, C_o \, D_{Li}^{1/2} \, v^{1/2}$$

where I_p is the peak current (A), n is the number of electrons transferred per molecule during the electrochemical reaction (for LTSO, n ~2), A is the active surface area of the electrode (cm^2), C_0 is the molar concentration of Li^+ in LTSO, D_{Li} is the Li^+ diffusion coefficient in LTSO ($cm^2 \cdot s^{-1}$), and v is the scanning rate ($V \cdot s^{-1}$). According to this formula, D_{Li} is proportional to $I_p^{-1/2}$.

S6. Electric conductivity and Li^+ diffusion coefficient at open circuit state[23]

According to the equivalent circuit as shown in the inset of **Figure** 2b, the Fitting results were listed in following Table:

Components	**R_s (ohm)**	**R_{ct} (ohm)**	**R_{total}^a (ohm)**
LTSO	2.8	169.5	172.3
3DC@LTSO	4.1	73.8	77.9

$^a R_{total}=R_s+R_{ct}$

The Li^+ diffusion coefficient at open circuit state could be calculated from the slanted lines in the Warburg region by Equation:

$$D_{Li} = R^2T^2/2A^2n^4F^4C_o^2\sigma^2$$

where D_{Li} is the Li^+ diffusion coefficient in LTSO ($cm^2 \cdot s^{-1}$), R is the gas constant (8.31 $J \cdot mol^{-1} \cdot K^{-1}$), T is the absolute temperature (298 K), A is the surface area of the cathode electrode (1.54 cm^2), n is the number of electrons transferred per molecule during the electrochemical reaction (for LTSO, n ~ 2), F is the Faraday constant (96485 $C \cdot mol^{-1}$), C_0 is the molar concentration of Li^+ in LTSO, and σ is the Warburg factor associated with Z_{Re} ($Z_{Re} \propto \sigma\omega^{-1/2}$)

Table S1. The elemental composition of LDAC of XPS and elemental analysis result

Name	Elemental composition (wt%) XPS result	Elemental composition (wt%) Elemental analysis result
C	94.2	92.5
O	5.80	3.85
N	-	0.45
H	-	2.30

Table S2. BET surface area of activated carbon with different ratio of KOH to the precursor

Number	BET surface area (m^2 g^{-1})	Ratio (KOH to the precursor)
1	1403	0.5:1
2	1871	1:1
3	2808	1.5:1
4	2017	2:1
5	1139	2.5:1

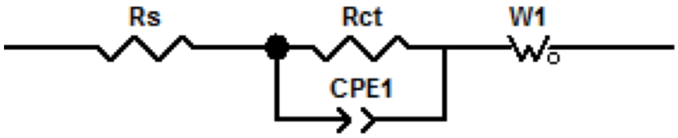

Figure S7. The electronic circuit equivalent of electrochemical impedance spectroscopies.

Tabls S3. The simulated R_s, R_{ct} and R_{ion} form equivalen circuit with the series of potential

Poteential (V)	R_s (ohm)	R_{ct} (ohm)	R_{ion} (ohm)
1.1	4.177	29.02	12.3
1.0	4.178	24.3	9.034
0.9	4.182	22.24	7.653
0.8	4.173	20.81	6.913
0.7	4.17	21.24	6.958
0.6	4.141	20.37	6.066
0.5	4.122	21.22	5.246
0.4	4.079	19.41	5.108
0.3	4.058	20.82	4.469
0.2	4.008	5.896	42.87
0.1	3.943	7.437	36.98
0	3.931	10.73	27.58
0.1	4.023	9.81	45.17
0.2	4.053	7.197	50.43
0.3	4.112	6.221	57.55
0.4	4.155	23.62	4.865
0.5	4.191	23.52	5.394
0.6	4.234	24.32	5.589
0.7	4.26	23.91	5.101
0.8	4.286	24.78	5.604
0.9	4.278	23.35	5.362
1.0	4.292	26.05	6.951
1.1	4.291	28.7	8.552

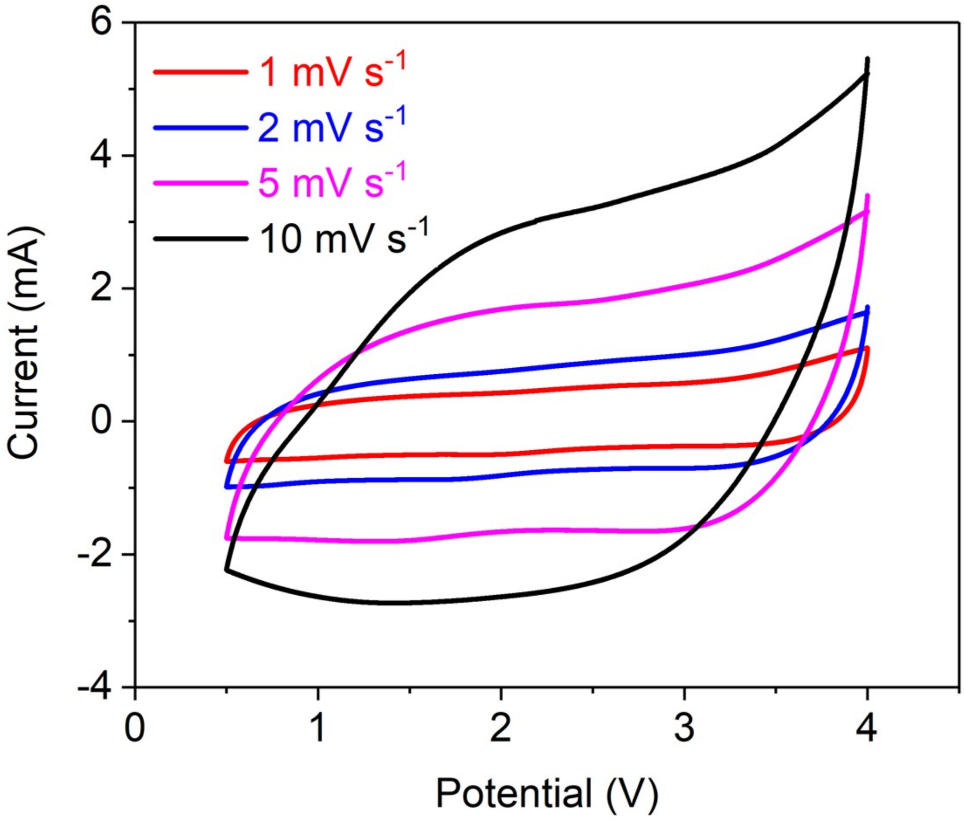

Figure S8. The CV curves of LDAC//3DC@LTSO LIC system with the voltage of 0.5~4.0V at the scanning rates of 1mV s^{-1}, 2mV s^{-1}, 5mV s^{-1} and 10mV s^{-1}.

Table S4. Detailed performance comparison of LTO and LTSO based LIC

System		Electrochemical performance	Reference
LDAC//3DC@LTSO	ED*	115.3 Wh kg^{-1} at 163.5 W kg^{-1}	This work
	PD*	6560 W kg^{-1} at 60 Wh kg^{-1}	
AC//G-LTO	ED	44 Wh kg^{-1} at 45 W kg^{-1}	Ref. 42
	PD	7200 W kg^{-1} at 11.4 Wh kg^{-1}	
AC//LTO	ED	30 Wh kg^{-1} at 60 W kg^{-1}	Ref. 44
	PD	1600 W kg^{-1} at 12 Wh kg^{-1}	
AC//C-LTO	ED	40 Wh kg^{-1} at 50 W kg^{-1}	Ref. 40
	PD	7500 W kg^{-1}at 21 Wh kg^{-1}	
AC//LTO+**6%G**	ED	63 Wh kg^{-1} at 45 W kg^{-1}	Ref. 39
	PD	2700 W kg^{-1} at 14 Wh kg^{-1}	
AC//Li-LTO	ED	67 Wh kg^{-1} at 500 W kg^{-1}	Ref. 43
	PD	8000 W kg^{-1} at 28.5 Wh kg^{-1}	
ZHTP//LTO	ED	69 Wh kg^{-1} at 500 W kg^{-1}	Ref. 46
	PD	4000 W kg^{-1} at 36 Wh kg^{-1}	
PGC//LTO	ED	55 Wh kg^{-1} at 686.9 W kg^{-1}	Ref. 45
	PD	6500 W kg^{-1} at 37 Wh kg^{-1}	
AC//S-TiO_2	ED	92.7 Wh kg^{-1} at 260 W kg^{-1}	Ref. 41
	PD	5000 W kg^{-1} at 52 Wh kg^{-1}	

ED*: Energy density; PD*: Power density