

## Electronic Supplementary Information

### A book-like organic based electrode with high areal capacity for high performance flexible lithium/sodium-ion batteries

#### Experimental section

##### Preparation of porous single layer electrode

Take 21 mg PTCDI (3,4,9,10-Perylenetetracarboxylic diimide) and 9 mg single walled carbon nanotubes and place them in a beaker. Then pour in 100ml of ethanol solution. The beaker was then ultrasound for 6 hours. At the end of the ultrasound, the mixed solution is pumped to form a membrane. The membrane was placed in the oven and vacuum dried at 70 °C for 12 hours.

##### Preparation of batteries

Areal specific capacity test using 2032 coin cells. Multiple single-layer electrodes are used directly for the cathode and no paste is required. The anode uses lithium or sodium metal.

The book-like electrodes are fabricated from single-layer porous flexible organic electrodes. Coat one side of the monolayer electrode with silver paste, then attach the other monolayer electrode, and repeat until the number of layers reaches the target number (Fig. S1e). Note that all monolayer electrodes can only be glued to the same side, otherwise the flexibility will be lost. Then, the multi-layer electrode was placed in an oven at 70 °C for 1 hour to form a thick, multi-layer, flexible book-like electrode.

The pouched pack battery fabrication uses the book-like flexible as cathode, carbon cloth embedded with lithium as anode.

##### Bending Angle calculation

Outside length =  $L_1$ , inner side length =  $L_2$ , the thickness of the electrode =  $d$ , bending angle =  $\theta$ .  
See fig. S4c for other parameters.

$$L_1 = 2l_1 + al_1, al_1 = 2\pi r \frac{\theta}{360^\circ}$$

$$L_2 = 2l_1 + al_2, al_2 = 2\pi(r + d) \frac{\theta}{360^\circ}$$

If the electrode does not break when bent, then

$$L_2 - L_1 \leq 0.094L_1$$

$$al_2 - al_1 \leq 0.094L_1$$

$$\frac{\pi d \theta}{180^\circ} \leq 0.094L_1$$

When the three-point doing bending test,  $L_1$  is 2 cm (instrument parameters).

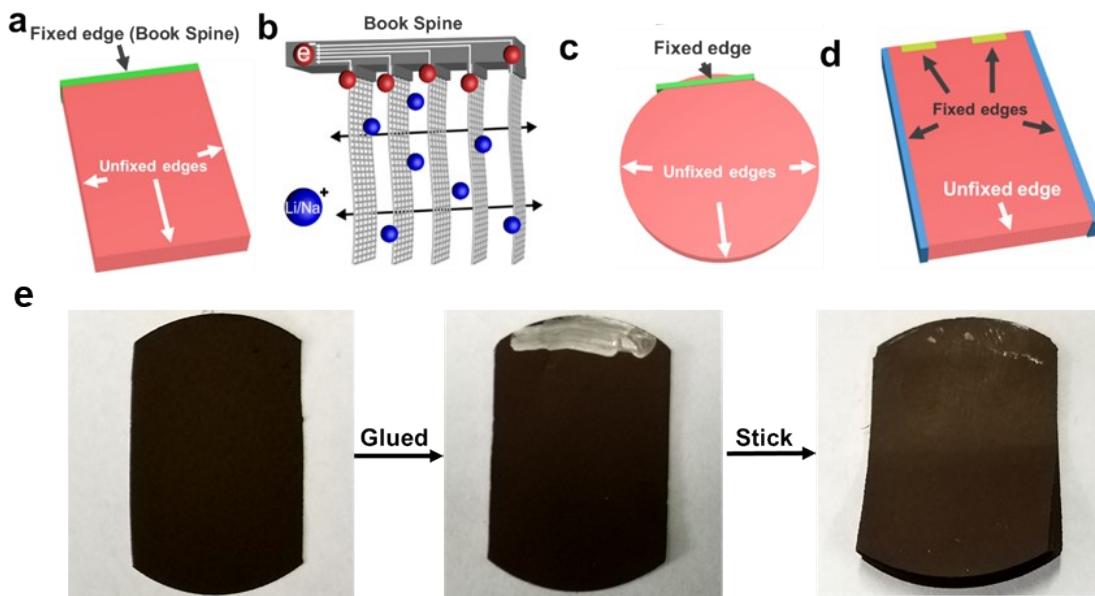
$$\frac{\pi d\theta}{180^\circ} \leq 0.188 \text{ cm}$$

$$d\theta \leq 0.188 \text{ cm} \times \frac{180^\circ}{\pi}$$

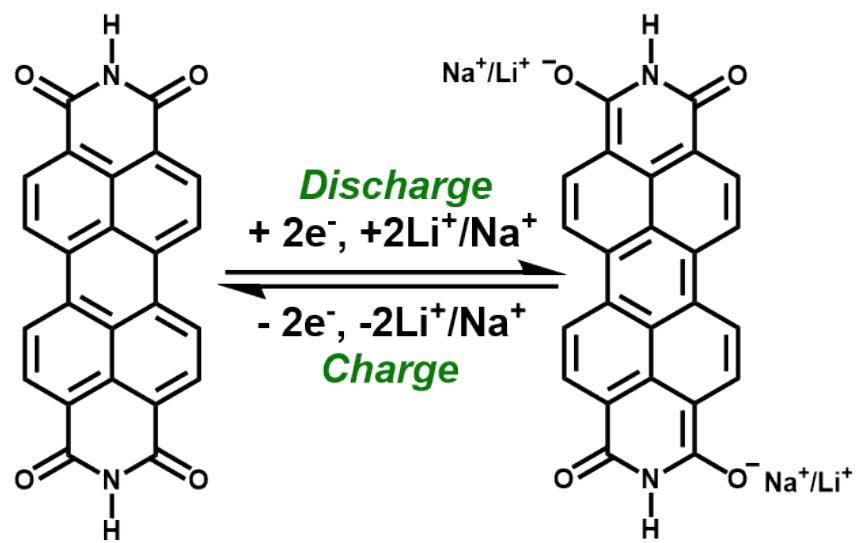
When  $\theta = 180^\circ$ ,  $d \leq 0.0598 \text{ cm}$

**Table S1** The areal capacity of the book-like electrode in flexible LIB & SIB is compared with other work.

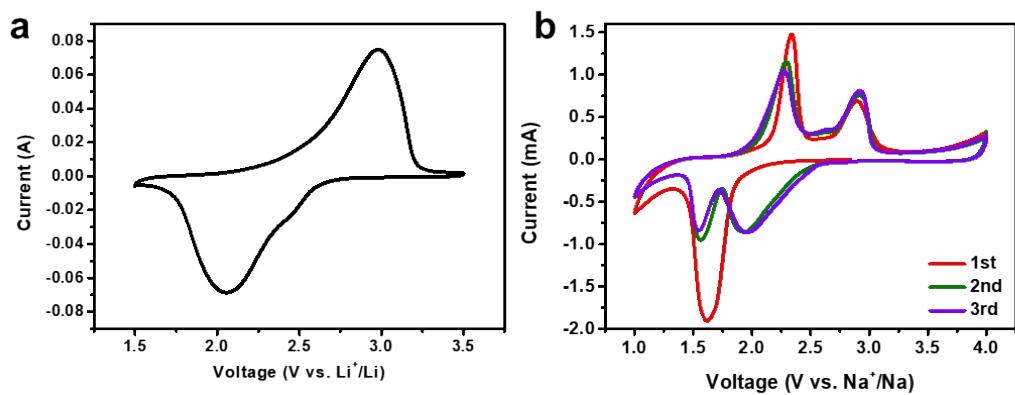
Published date	Current density ( $\text{mA g}^{-1}$ )	Areal capacity ( $\text{mAh cm}^{-2}$ )	Materials	Flexible Batteries	Journal	Ref.
2022/2/6	83.75	5.2	O-Ti <sub>3</sub> C <sub>2</sub> @CNF	LIB	<i>Energy Stor. Mater.</i>	<sup>1</sup>
2021/3/3	17	4.56	LFP/CNT/EVA	LIB	<i>Adv. Funct. Mater</i>	<sup>2</sup>
2020/4/1	98	4.78	P@MOF/CNT/CFC	LIB	<i>Chem. Eng. J.</i>	<sup>3</sup>
2019/11/4	28.2	2.01	LiMn <sub>2</sub> O <sub>4</sub> @GCN	LIB	<i>Nano Energy</i>	<sup>4</sup>
2017/7/31	100	0.56	V2O5@polydopamine@carbon cloth	LIB	<i>J. Power Sources</i>	<sup>5</sup>
2017/2/27	138	1.4	N-doped carbon@LiTi <sub>2</sub> (PO <sub>4</sub> ) <sub>3</sub>	LIB	<i>J. Mater. Chem. A</i>	<sup>6</sup>
2016/2/10	37.2	0.25	3D CNTs	LIB	<i>Nanotechnoloy</i>	<sup>7</sup>
2021/8/24	20	4.5	TiO <sub>2</sub> -C	SIB	<i>Infomat</i>	<sup>8</sup>
2020/5/20	40	2.45	3D porous graphene nanosheet/SnS <sub>2</sub>	SIB	<i>Small</i>	<sup>9</sup>
2019/11/26	32	0.76	K <sub>2</sub> Zn <sub>3</sub> (Fe(CN) <sub>6</sub> ) <sub>2</sub> ·9H <sub>2</sub> O@carbon cloth	SIB	<i>Small</i>	<sup>10</sup>
2019/1/7	83	2.12	3D Carbon-Networks/Fe <sub>7</sub> S <sub>8</sub> /Graphene	SIB	<i>Adv. Mater.</i>	<sup>11</sup>
<b>Our work</b>	100	5.88	Book-like PTCDA@CNTs	LIB		
	27.32	5.24		SIB		



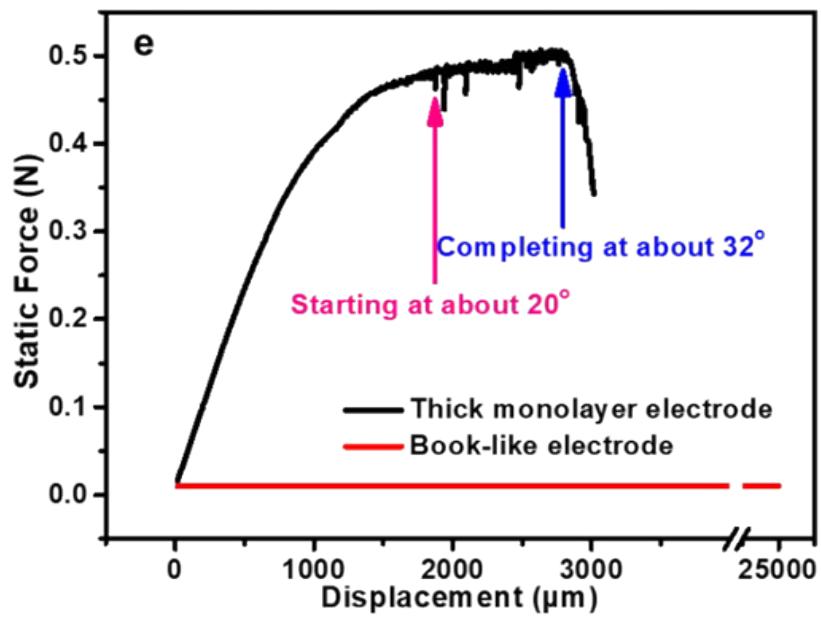
**Fig. S1** (a) Fixed edge and three free edges of the book-like electrode. (b) The movement of electrons on the book-like electrode and the movement of lithium ions or sodium ions between the porous multilayer electrode in the radial direction. (c) The fabrication method of book-like electrode is applied to the flexible circular multilayer electrode. (d) A conventional cathode-anode alternating stack cell, with the sides fixed by a zigzag separator (blue) and the head fixed by lugs welding (yellow). (e) Pictures of the book-like electrode made of conductive silver paste bonded layers.



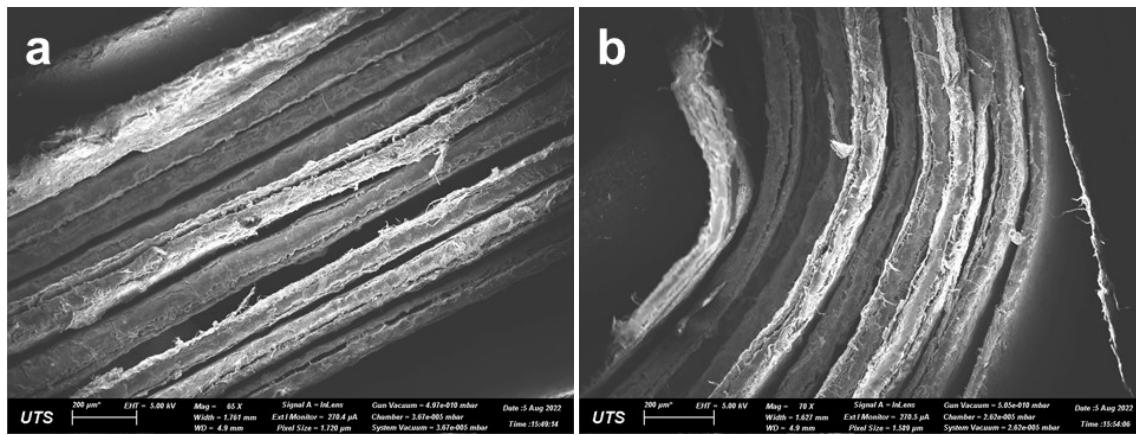
**Fig. S2** Two-electron REDOX reaction mechanism of PTCDA in LIB & SIB.



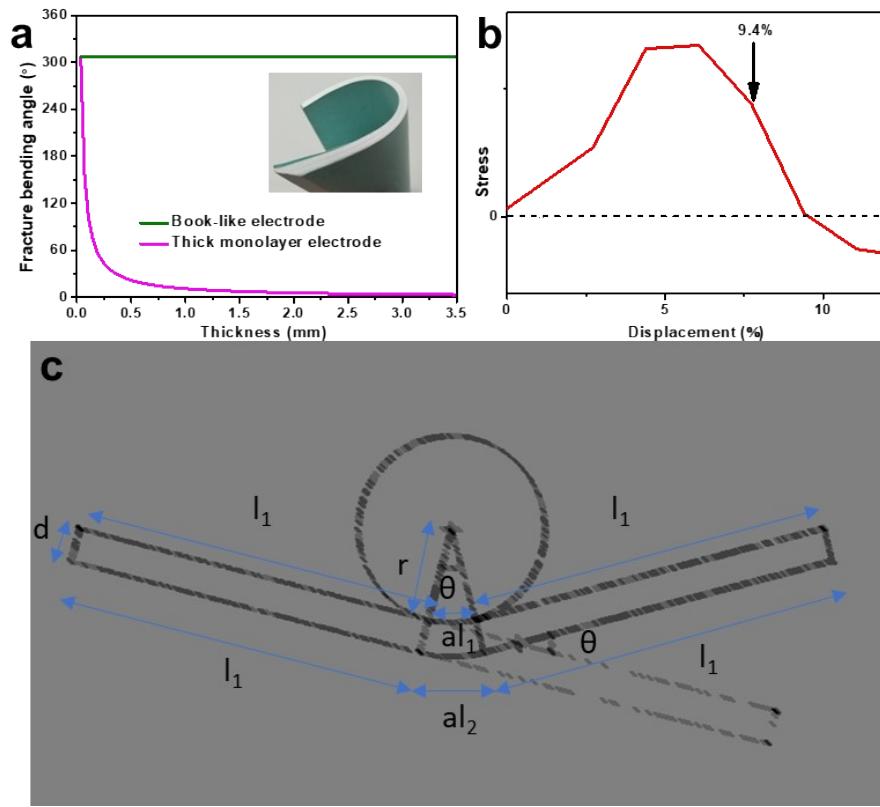
**Fig. S3** CV of book-like electrode in LIB (a) & SIB (b).



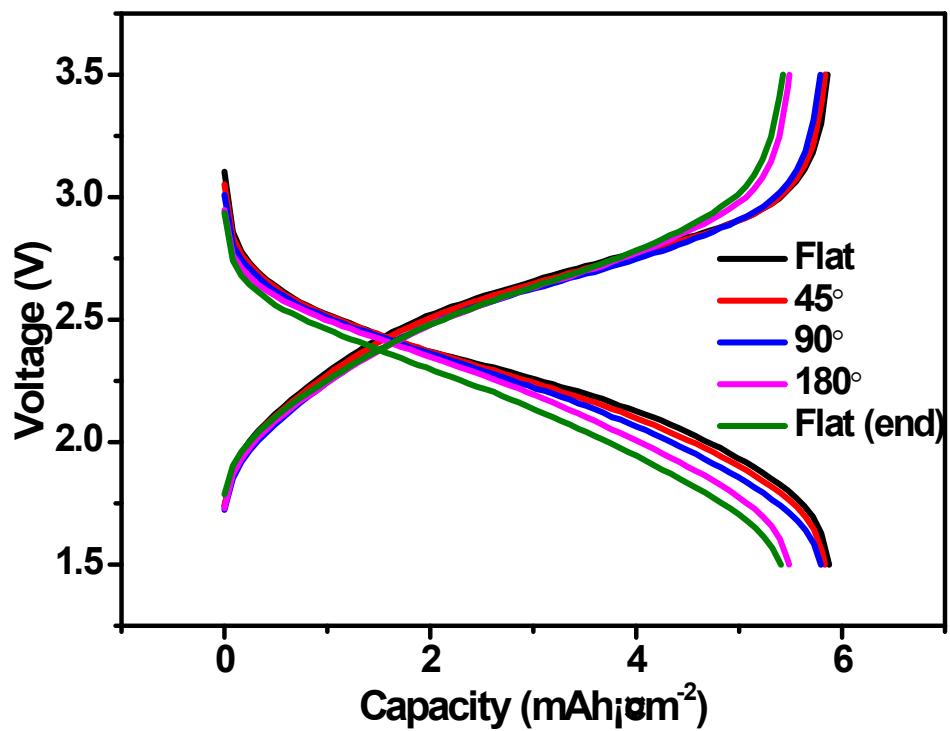
**Fig. S4** Force curves of two electrodes in three-point bending test.



**Fig. S5** The book-like electrode cross-section SEM images before (a) after the bending test (b)



**Fig. S6** (a) The fracture bending angles of electrodes with different thickness. (b) Tensile fracture stress-strain diagram of the electrode. (c) Length parameters of each part when the electrode is bent.



**Fig. S7** Galvanostatic charge-discharge curves of flexible book electrode in bending test of lithium-ion battery



**Fig. S8** From left to right are photos of flexible batteries applied in a smart insole and wearable LED arrays.

## References

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