Supporting Information

High-Capacity Ultra-Thin Flexible Lithium-Ion Batteries with Enhanced Rate Capability by a Cast All-in-One Cathode-Separator-Anode Monolith †

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Fig. S1 Photographs of PVDF solutions in various solvents: (a-c) after stirring for 24 hours and (d-f) after an additional 24 hours of resting. The solvents used are: (a, d) Acetone only, (b, e) NMP only, and (c,f) acetone-NMP (acetone:NMP = 6:1 v/v).



Fig. S2 (a)–(d) SEM images of cathode-separator-anode monoliths and (e)–(h) corresponding EDS elemental mapping (yellow: Ti, light-green: C, light-blue: Ni, pink: Co).



Fig. S3 (a) Photographs of the NCM-Gra cathode-separator-anode monolith in folded and unfolded condition. The red arrow points to the folded line. (b) Photographs of the NCM-Gra cathode-separator-anode monolith electrode in a rolled state.



Fig. S4 A photograph and SEM images of NCM-Gra cathode-separator-anode monolith after folding and unfolding. (a) The SEM image (1) corresponds to the area within the orange circle, and (2) corresponds to the area within the red circle in the photograph. (b) Higher magnifications of (2) in (a).



Fig. S5 (a) Conformation of PVDF chains in solvents: NMP, acetone, and an acetone-NMP mixture. (b) Behavior of PVDF chains under bending and releasing when processed in the acetone-NMP mixture.



Fig. S6 Galvanostatic charge–discharge curves of coin type cells at different rates between 0.2 and 10 C with (a) a single all-in-one cathode-separator-anode monolith, (b) freestanding electrodes, and (c) conventional electrodes.



Fig. S7 Cycle performance of coin-type full cells with a single all-in-one-monolith at 1 C.

Ref.	Monolithic cell structure	Thickness	Arial capacity (mAh cm ⁻²)	Max. discharge rate (coin cell)	Nominal discharge voltage (V)	Flexibility	Absence of metallic current collector
this work	Ο	0	4.9	10 C	3.8 V	Ο	0
[1]	0	N/A	1.3	1C	N/A	\triangle	Х
[2]	0	0	0.005	5C	N/A	\triangle	Х
[3]	0	Х	N/A	8.3 C	1.7 V	0	0
[4]	0	0	3	3C	2.3 V	\triangle	0
[5]	0	0	0.67	10 C	1.2 V	0	0
[6]	0	Х	N/A	2C	1.8V	0	0
[7]	Х	Х	0.8	1C	3.4 V	0	0
[8]	Х	Х	5.6	3 C	3.6 V	0	Х
[9]	Х	0	4.5	1.8 C	3.8 V	0	Х

• 'O' in 'Thickness' means the thickness of pouch cell is under 1 mm; 'X' means the thickness is over 1 mm.

• 'O' in 'Flexibility' means the 'very flexible that can fold-unfold or twist'; '△' means the flexibility is 'not flexible enough but can bend'.

Table S1. Summary of the performance of flexible cells with monolithic and non-monolithic electrodes.

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