Electronic Supplementary Information

Conversion pseudocapacitance-contributing and robust heteronanostructural perovskite KCo_{0.54}Mn_{0.46}F₃ nanocrystals anchored on graphene nanosheets anodes for advanced lithium-ion capacitor, battery and their hybrids

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Experimental section

Synthesis of materials

The chemicals and reagents used in this paper are of analytical level (A.R.) and are used without further treatment (Table S1). A facile one-pot solvothermal method is applied to synthesize these perovskite fluorides. As all the synthetic steps of KCMF are the same except for the usage amounts of cobalt and manganese salts, the synthetic procedure of KCMF(3:2) is taken out as an example: i) Dissolve 1.2 mmol CoCl₂·6H₂O, 0.8 mmol MnCl₂·4H₂O, 5 mmol KF·2H₂O and 0.2 g PVP-K30 into 35 mL solvents of ethylene glycol (EG); ii) Magnetically stir the mixture thoroughly and disperse it in an ultrasonic bath at 100 W for 30 minutes; iii) transfer the mixture into a 50 mL Teflon-lined stainless steel autoclave, heat it at 180°C for 12 h in an electric oven, then cool down naturally; iv) Centrifuge the yielded precipitates with absolute alcohol for several times to wash the product; v) Dry it at 95 °C for 12 h to obtain the final product. The synthesis procedure of U-KCMF(3:2)/rGO is the same as KCMF(3:2), except that 50 mg graphene oxides (GO) are dispersed in 35mL EG with the ultrasonic treatment for 3 h before the raw materials adding into the solvents. The KCMF(3:2)/rGO sample is obtained by ball-milling the U-KCMF(3:2)/rGO powder under the condition of 1500 rpm for 1 h.

Characterizations

The phases and crystallinity properties are determined by X-ray diffraction (XRD). The surface chemical compositions and electronic structures are checked by X-ray photoelectron spectra (XPS). The morphology and size of particles are analyzed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The crystalline microstructures are resolved by the high-resolution TEM (HRTEM) and selected area electron diffraction (SAED). The element composition and distribution are measured by inductively coupled plasma-optical emission spectrometer (ICP-OES), X-Ray energy dispersive spectra (EDS) and mapping. The specific surface area, pore volume and size distribution are examined by nitrogen isothermal sorptions with Brunauer-Emmett-Teller (BET) and Barrett-Joyner-Halenda (BJH) methods. The existence and content of reduction graphene oxide are analyzed by Raman spectrum (RS) and Thermogravimetric analysis (TG).

Electrochemical measurements

The following is the procedure of preparing electrodes: i) Mix 70 wt.% active materials (as-synthesized KCMF, U-KCMF(3:2)/rGO, KCMF(3:2)/rGO or commercial AC, LFP, AC/LFP (3:1, 1:1, 1:3 in weight)), 20 wt.% conductive agent of acetylene black (AB) and 10 wt.% binder of polyvinylidene fluoride (PVDF) evenly with N-methyl-2pyrrolidone (NMP)); ii) Caste the mixture onto Cu foil and carbon-coated Al foil used for the current collector of anodes and cathodes respectively; iii) Dry them in a vacuum oven at 110 °C for 12 h; iv) Cut them into disks with diameter of 12 mm with the active materials mass loading of about 1.2-5 mg cm⁻². In this paper, half-cells are fabricated using the 2032 coin cells with active materials as working electrodes, a Li plate as both counter and reference electrodes, glass fiber as separator and 1 M LiPF₆ dissolving in the mixed solvents of ethylene carbonate (EC), ethyl methyl carbonate (EMC) and dimethyl carbonate (DMC) (1:1:1 in volume) with 1% vinylene carbonate (VC) additives (LBC-305-01, CAPCHEM) as electrolyte to test the performance of materials. Tests for the different energy storage devices (LICs, LIBs, LIC/Bs) are conducted via full-cells with certain mass ratios of anode and cathode active materials (Table S6). All cell assemblies are performed in a high pure Ar-filled dry glovebox (MIKROUNA, O_2 and $H_2O<0.1$ ppm) and all tests are carried out at room temperature (about 25 °C) except the assigned tests under high (40 °C) and low (-20 °C) temperatures. (The more detailed information of the above-mentioned material, chemicals and reagents can be seen in Table S1)

Methods: calculations for m_+/m_- , C_m , E_m , P_m

The charge-balance (Q+=Q-) as shown in equation S(1) is used to calculated the mass ratios of cathodes and anodes materials. The specific capacity (C_m , mAh g⁻¹), energy density ($E_{m,1}$, Wh kg⁻¹) for LICs, energy density ($E_{m,2}$, Wh kg⁻¹) for LIBs and LIC/Bs, and power density (P_m , kW kg⁻¹) of the different energy storage devices are calculated according to the equations S(2), S(3), S(4) and S(5).

$$m_{+}/m_{-}=Q_{m-}/Q_{m+}$$
 S(1)

$$C_{\rm m} = Q_{\rm m}/3.6 = It/3.6m$$
 S(2)

$$E_{m,1}=(C_m \triangle V)/2$$
 S(3)

$$E_{m,2}=(C_mV)$$
 S(4)

$$P_{\rm m} = 3.6 E_{\rm m} / t_{\rm d}$$
 S(5)

Where m, Q_m , ΔV , V, I and t refer to the mass of active materials (g, for half cells, it means the mass of active materials of anode or cathode; for full-cells, it means the total masses of active materials of anode and cathode), specific charge or discharge quantity (C g⁻¹, for anode, it means the charge quantity; for cathode and full-cells, it refers to the discharge quantity), potential window (V), potential of the discharging plateaus (V), current (A) and discharging time (s), respectively.

Supplemental figures





(b)

Sample	ICDD-PDF	Crystal system	Space group	Cell (a×b×c)/Å ³
KC0F3	18-1006	Cubic	Pm-3m	4.0708×4.0708×4.0708
KMnF3	17-0116	Cubic	Pm-3m	4.1890×4.1890×4.1890

Figure S1. The crystalline structures of perovskite KMF_3 (a) and crystalline parameters for $KCoF_3$ and $KMnF_3$ (b).







Figure S3. SEM (a, b) and TEM (c, d) images of KCMF(3:2) sample.



Figure S4. EDS and mapping of KCMF(3:2) sample with a scale bar of 5 μ m.



Figure S5. EDS and mapping of KCMF(3:2) sample with a scale bar of 700 nm.



Figure S6. SEM (a, b), TEM (c) and HRTEM (d) images of U-KCMF(3:2)/rGO sample.



Figure S7. EDS and mapping of U-KCMF(3:2)/rGO sample with a scale bar of 1 μ m.



Figure S8. SEM (a, b) and TEM (c, d) images of KCMF(3:2)/rGO sample.



Figure S9. EDS and mapping of KCMF(3:2)/rGO sample with a scale bar of 600 nm.



Figure S10. RS of U-KCMF(3:2)/rGO and KCMF(3:2)/rGO samples (a) and TG of KCMF(3:2), U-KCMF(3:2)/rGO and KCMF(3:2)/rGO samples (b).



Figure S11. CV plots for the first three cycles of KCMF (6:1), (3:1), (1:1), (2:3), (1:3) and (1:6) electrodes at 0.3 mV s⁻¹.



Figure S12. GCD curves for the first five cycles of KCMF (Co/Mn=1:0 0 :1) electrodes at 0.1 A g⁻¹.



Figure S13. GCD curves for the respective 3^{rd} cycles at 3.2~0.1 A g⁻¹ of KCMF (Co/Mn=1:0~0:1) electrodes.



Figure S14. Rate performance and coulombic efficiency of KCMF (Co/Mn=1:0~0:1) electrodes at 0.1~3.2~0.1 A g⁻¹.



Figure S15. Cycling stability and coulombic efficiency of KCMF (Co/Mn=1:0~0:1) electrodes at 2 A $g^{\rm -1}$ for 1000 cycles.



Figure S16. GCD curves for the first five cycles at 0.1 A g^{-1} (a, e), GCD curves for the respective 3^{rd} cycle at 3.2~0.1 A g^{-1} (b, f), rate performance and coulombic efficiency at 0.1~3.2~0.1 A g^{-1} (c, g), and cycling stability and coulombic efficiency at 2 A g^{-1} for 1000 cycles (d, h) of U-KCMF(3:2)/rGO and KCMF(3:2)/rGO electrodes.



Figure S17. The equivalent circuits used to fit the experimental Nyquist plots before (a) and after (b) cycling of KCMF(3:2), U-KCMF(3:2)/rGO and KCMF(3:2)/rGO electrodes.



Figure S18. CV plots for the first three cycles of KCMF(1:0) and (0:1) electrodes at 0.3 mV s⁻¹.



Figure S19. The comparison of CV plots for the 3^{rd} cycle at 0.1 and 0.3 mV s⁻¹ of KCMF(1:0) (a), (0:1) (b), (3:2) (c) U-KCMF(3:2)/rGO (d) and KCMF(3:2)/rGO (e) electrodes.



Figure S20. EDS (a), and mapping (b-g) for the KCMF(3:2) electrode discharged-0.01V state of the first GCD (0.05 A g^{-1}) cycle.



Figure S21. EDS (a), and mapping (b-g) for the KCMF(3:2) electrode charged-3.0V state of the first GCD (0.05 A g^{-1}) cycle.



(b)

Phases	PDF Card	Crystal system	Space group	Cell (a×b×c)/Å ³
Со	05-0727	Hexagonal	P63/mmc	2.503×2.503×4.061
Mn	17-0910	Tetragonal	I4/mmm	2.672×2.672×3.550
CoF ₂	38-0883	Cubic	Pa-3	4.958×4.958×4.958
MnF ₂	34-1326	Tetragonal	P-42m	5.112×5.112×5.256
LiF	45-1460	Cubic	Fm-3m	4.027×4.027×4.027
KF	36-1458	Cubic	Fm-3m	5.348×5.348×5.348
Li ₂ CO ₃	22-1141	Monoclinic	C2/c (15)	8.359×4.977×6.194

Figure S22. Schematics of reaction mechanisms for KCMF(3:2) electrode during the discharging/charging processes under the first two cycles (a); Crystalline structure information of Co, Mn, CoF₂, MnF₂, LiF, KF and Li₂CO₃ phases (b).

(a)



Figure S23. Rate performance and coulombic efficiency at 0.1~3.2~0.1 A g⁻¹ (a), cycling stability and coulombic efficiency at 1 A g⁻¹ for 1000 cycles (b), CV plots for the first three cycles at 10 mV s⁻¹ (c), GCD curves for the first five cycles at 0.1 A g⁻¹ (d) and GCD curves for the respective 3rd cycles at 3.2~0.1 A g⁻¹ (e) of AC electrode.



Figure S24. Rate performance and coulombic efficiency at $0.1^{-3}.2^{-0.1}$ A g⁻¹ (a), cycling stability and coulombic efficiency at 1 A g⁻¹ for 1000 cycles (b), CV plots for the first three cycles at 10 mV s⁻¹ (c), GCD curves for the first five cycles at 0.1 A g⁻¹ (d) and GCD curves for the respective 3rd cycles at 3.2~0.1 A g⁻¹ (e) of LFP electrode.



Figure S25. The comparisons of CV plots at 10 mV s⁻¹ (a), specific capacity and coulombic efficiency derived from the respective 3^{rd} cycle at 3.2-0.1 A g⁻¹ (b), GCD curves for the 3^{rd} cycle at 0.1 A g⁻¹ (c) and cycling stability for 1000 cycles at 1 A g⁻¹ (d) between AC/LFP(3:1), (1:1) and (1:3) electrodes.



Figure S26. Rate performance and coulombic efficiency at $0.1^{-3}.2^{-0.1}$ A g⁻¹ (a), cycling stability and coulombic efficiency at 1 A g⁻¹ for 1000 cycles (b), CV plots for the first three cycles at 10 mV s⁻¹ (c), GCD curves for the first five cycles at 0.1 A g⁻¹ (d) and GCD curves for the respective 3rd cycles at 3.2^{-0.1} A g⁻¹ (e) of AC/LFP(3:1) electrode.



Figure S27. Rate performance and coulombic efficiency at 0.1~3.2~0.1 A g⁻¹ (a), cycling stability and coulombic efficiency at 1 A g⁻¹ for 1000 cycles (b), CV plots for the first three cycles at 10 mV s⁻¹ (c), GCD curves for the first five cycles at 0.1 A g⁻¹ (d) and GCD curves for the respective 3rd cycles at 3.2~0.1 A g⁻¹ (e) of AC/LFP(1:1) electrode.



Figure S28. Rate performance and coulombic efficiency at $0.1^{-3}.2^{-0.1}$ A g⁻¹ (a), cycling stability and coulombic efficiency at 1 A g⁻¹ for 1000 cycles (b), CV plots for the first three cycles at 10 mV s⁻¹ (c), GCD curves for the first five cycles at 0.1 A g⁻¹ (d) and GCD curves for the respective 3rd cycles at 3.2^{-0.1} A g⁻¹ (e) of AC/LFP(1:3) electrode.



Figure S29. CV windows at 30 mV s⁻¹ of KCMF(3:2)//AC, U-KCMF(3:2)/rGO//AC and KCMF(3:2)/rGO//AC LICs (a, d, g); KCMF(3:2)//LFP, U-KCMF(3:2)/rGO//LFP and KCMF(3:2)/rGO//LFP LIBs (b, e, h); KCMF(3:2)//AC/LFP(1:3), U-KCMF(3:2)/rGO//AC/LFP(1:3) and KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs (c, f, i).



Figure S30. CV plots at 10~160 mV s⁻¹, GCD curves at 0.5~16 A g⁻¹, rate performance and coulombic efficiency at 0.5~16 A g⁻¹ and cycling stability and coulombic efficiency at 5 A g⁻¹ for 5000 cycles of KCMF(3:2)//AC LICs under the working voltages of 0~4.0 V (a-d), 0~4.4 V (e-h), U-KCMF(3:2)/rGO//AC LICs under the working voltages of 0~4.0 V (i-l), 0~4.4 V (m-p) and KCMF(3:2)/rGO//AC LICs under the working voltages of 0~4.0 V (q-t), 0~4.4 V (u-x).



Figure S31. CV plots at 10~160 mV s⁻¹, GCD curves at 0.5~16 A g⁻¹, rate performance and coulombic efficiency at 0.5~16 A g⁻¹ and cycling stability and coulombic efficiency at 5 A g⁻¹ for 5000 cycles under the working voltage of 0~4.7 V of KCMF(3:2)//LFP LIBs (a-d), U-KCMF(3:2)/rGO//LFP LIBs (e-h) and KCMF(3:2)/rGO//LFP LIBs (i-l).



Figure S32. CV plots at 10~160 mV s⁻¹, GCD curves at 0.5~16 A g⁻¹, rate performance and coulombic efficiency at 0.5~16 A g⁻¹ and cycling stability and coulombic efficiency at 5 A g⁻¹ for 5000 cycles of KCMF(3:2)//AC/LFP(1:3) LIC/Bs under the working voltages of 0~4.4 V (a-d), 0~4.7 V (e-h), U-KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs under the working voltages of 0~4.4 V (i-l), 0~4.7 V (m-p) and KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs under the working voltages of 0~4.4 V (q-t), 0~4.7 V (u-x).



Figure S33. The performance of KCMF(3:2)/rGO//AC LICs under high (40 °C) and low (-20 °C) temperature: CV windows at 10 mV s⁻¹ (a, g), CV plots at 10-160 mV s⁻¹ (b, h), rate performance (c, i), GCD curves (d, j), Ragone plots (e, k) and cycling behavior at 2 A g⁻¹ for 5000 cycles (f, l) under the working voltage of 0-4.0 V.



Figure S34. The performance of KCMF(3:2)/rGO//LFP LIBs under high (40°C) and low (-20°C) temperature: CV windows at 10 mV s⁻¹ (a, g), CV plots at 10-160 mV s⁻¹ (b, h), rate performance (c, i), GCD curves (d, j), Ragone plots (e, k) and cycling behavior at 2 A g⁻¹ for 5000 cycles (f, l) under the working voltage of 0-4.7 V.



Figure S35. The performance of KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs under high (40°C) and low (-20°C) temperature: CV windows at 10 mV s⁻¹ (a, g), CV plots at 10-160 mV s⁻¹ (b, h), rate performance (c, i), GCD curves (d, j), Ragone plots (e, k) and cycling behavior at 2 A g⁻¹ for 5000 cycles (f, l) under the working voltage of 0-4.4 V.

Supplemental tables

Materials, chemicals and reagents	Туре	Company	Characteristics
CoCl ₂ ·6H ₂ O	AR	SinoPharm	Purity≥99.0%
MnCl ₂ ·4H ₂ O	AR	SinoPharm	Purity≥99.0%
KF·2H ₂ O	AR	SinoPharm	Purity≥99.0%
PVP-K30	GR	SinoPharm	Purity _{299.8%}
EG	AR	SinoPharm	Purity≥99.0%
Graphene oxide (GO)	Powder	ZhengZhou JingHong New Energy	Purity≥94.0% 3-10 floors Conductivity: 2.0*10 ⁵ S m ⁻¹ SSA: 150~200 m ² g ⁻¹
AC	YEC 8b	FuZhou YiHuan	D50: ~10 μm; Density: 0.4 g cm ⁻³ ; SSA: 2000~2500 m ² g ⁻¹
LiFePO ₄	LFP-NCO	Aleees	D50: $4 \pm 2 \mu m$; Tab: $1 \pm 0.2 \text{ g cm}^{-3}$; SSA: $13 \pm 2 \text{ m}^2 \text{ g}^{-1}$
AB	Battery grade	#	#
NMP	AR	Kermel	Purity≥99.0%
PVDF	Battery grade	#	#
Electrolytes	LBC-305-01	CAPCHEM	1 M LiPF ₆ /EC: EMC: DMC (1:1:1)/1% VC
Li plate	15.6*0.45 mm	China Energy	15.6*0.45 mm
Cu foil	200*0.015	GuangZhou JiaYuan	Total thickness: 15 μm; Weight: 87 g m ⁻²
Carbon coated-Al foil	222*0.015	GuagZhou NaNuo	Total thickness: 17 μm; Strength: 192 Mpa
Glass microfiber filters	GF/D 2.7 μm 1823-025	Whatman	Diameter: 25 mm; Thickness: 675 µm; Weight: 121 g m ⁻²
Cell components	CR-2032	ShenZhen TianChenHe	#

 Table S1. Materials, chemicals and reagents used in this study.

Sample	Element	Wt.%	The final chemical composition
	K	24.99	
VCME(2,2)	Со	21.40	K C. M. F
KCMF(3:2)	Mn	16.41	K0.97C00.55IVIN0.45F2.96
	F	37.20	
(b)			
Sample	Element	Wt.%	The final chemical composition
	K	18.70	
U-KCMF(3:2)/rGO	Со	16.84	$K_{0.93}Co_{0.54}Mn_{0.46}F_{3.0}$
	Mn	12.89	
(c)			
Sample	Element	Wt.%	The final chemical composition
	К	18.20	
KCMF(3:2)/rGO	Со	14.45	$K_{1.02}Co_{0.54}Mn_{0.46}F_{3.0}$
	Mn	11.52	

Table S2. ICP results of KCMF(3:2) (a), U-KCMF(3:2)/rGO (b) and KCMF(3:2)/rGO (c) samples.

(a)

	Specific capacity/(mAh g ⁻¹)										
<i>i</i> /(A g ⁻¹)	KCMF electrodes (Co/Mn=1:0~0:1)										
	1:0	6:1	3:1	3:2	1:1	2:3	1:3	1:6	0:1	U-(3:2)/rGU	(3:2)/rGU
0.1	144	109	108	126	108	101	88	88	76	339	288
0.2	123	91	89	106	92	87	79	77	71	288	259
0.4	98	72	75	87	77	75	71	68	64	230	215
0.8	71	58	62	71	64	63	61	60	55	180	177
1.6	51	44	48	56	51	52	51	48	45	134	138
3.2	36	32	36	41	39	40	40	38	36	92	106
Cycling behavior retention%/2 A g ^{-1/} 1000 cycles	190	207	213	232	175	188	187	182	183	125	169

Table S3. Specific capacity of KCMF (Co/Mn=1:0~0:1), U-KCMF(3:2)/rGO and KCMF(3:2)/rGO electrodes.

	Specific capacity (mAh g ⁻¹)						
<i>i</i> /(A g ⁻¹)	KCo _{0.54} Mn _{0.46} F3/rGO This work	KNi_{0.1}Co_{0.9}F₃ [1]. J. Mater. Chem. A, 2019, 7 , 8315-8326	Na _{0.85} Ni _{0.45} Co _{0.55} F _{3.56} [2]. Chem. Commun., 2019, 55 , 6739-6742				
0.1	288	166	286				
0.2	259	161	250				
0.4	215	147	219				
0.8	177	124	184				
1.6	138	99	149				
3.2	106	73	115				
Cycling behavior retention	169%/1000 cycles/ 2 A g ⁻¹	197%/1000 cycles/1 A g ⁻¹	187%/600 cycles/1 A g ⁻¹				

Table	S4.	А	comparison	for	reported	perovskite	fluorides	anodes	for	lithium-ion
storag	e.									

EIS parameters						
Before cycling	KCMF(3:2)	U-KCMF(3:2)/rGO	KCMF(3:2)/rGO			
Model		R(QR)W(QR)				
$R_{\rm s}\left(\Omega ight)$	3.346	5.074	3.173			
Q_1 (S·sec ⁿ)	1.556×10-5	2.045×10-5	2.7×10 ⁻⁵			
\mathbf{n}_1	0.8554	0.8288	0.8069			
$R_{ m ct}\left(\Omega ight)$	63.75	49.63	47.21			
$W(\mathbf{S}\cdot\mathbf{sec}^{0.5})$	0.01631	0.009539	0.008319			
$Q_2(\mathbf{S}\cdot\mathbf{sec}^n)$	5.951×10 ⁻⁴	9.446×10 ⁻⁴	1.183×10 ⁻³			
n ₂	0.9738	0.9715	0.9629			
$R_{ m e}\left(\Omega ight)$	6.984×10 ⁵	3.305×10 ⁵	3.127×10 ⁵			
χ^2	3.049×10 ⁻³	2.461×10 ⁻³	2.82×10 ⁻³			

Table S5. EIS parameters of KCMF(3:2), U-KCMF(3:2)/rGO and KCMF(3:2)/rGO electrodes before (a) and after (b) cycling.

(<u>b</u>)

(a)

EIS parameters							
After cycling	KCMF(3:2)	U-KCMF(3:2)/rGO	KCMF(3:2)/rGO				
Model							
$R_{\rm s}\left(\Omega ight)$	4.451	6.549	7.972				
<i>C</i> _{dl, 1} (F)	5.965×10-6	2.494×10 ⁻⁵	1.257×10 ⁻⁵				
$R_{\mathrm{SEI}}\left(\Omega ight)$	2.66	3.507	4.429				
C _{dl, 2} (F)	7.51×10 ⁻³	2.983×10 ⁻⁶	4.747×10 ⁻³				
$R_{\mathrm{ct},1}\left(\Omega\right)$	6.086	4.934	8.754				
Q (S·sec ⁿ)	1.564×10 ⁻⁴	2.242×10 ⁻³	1.05×10 ⁻⁴				
n	0.7445	0.5587	0.7648				
$R_{\rm ct,2}(\Omega)$	36.21	29.66	49.36				
$W(\mathbf{S}\cdot\mathbf{sec}^{0.5})$	0.02694	0.04528	0.01931				
$C_{\rm int}({\rm F})$	0.01972	0.08658	0.02547				
χ^2	7.584×10-4	3.183×10 ⁻⁴	3.914×10-4				

<i>i</i> /(A g-1)	Specific capacity/(mAh g ⁻¹)						
<i>U</i> (A g ⁻)	AC	LFP	AC/LFP(3:1)	AC/LFP(1:1)	AC/LFP(1:3)		
0.1	77	145	83	97	125		
0.2	72	139	78	89	119		
0.4	65	129	71	79	109		
0.8	58	115	65	68	99		
1.6	50	100	56	57	86		
3.2	43	83	48	48	73		
Cycling behavior retention%/1 A g ⁻¹ / 1000 cycles	71	92	78	89	89		

Table S6. Specific capacity of positive electrodes (AC, LFP, AC/LFP(3:1), AC/LFP(1:1) and AC/LFP(1:3)).

		$Q_{\rm m}$ (mAh g ⁻¹ , 0.1A/g)						
Energy storage	Name	Р	ositive el	ectrode	Ν	Negative electrode		
devices		AC	LFP	AC/LFP (1:3)	KCMF (3·2)	U-KCMF (3·2)/rGO	KCMF (3·2)/rGO	<i>m</i> ₊ / <i>m</i> ₋
	KCMF(3:2) //AC	77		(110)	126	(012)/100	(0.2)/100	1.6:1
Lithium-ion capacitors (LICs)	U-KCMF(3:2)/rGO //AC	77				339		4.4:1
	KCMF(3:2)/rGO //AC	77					288	3.7:1
	KCMF(3:2) //LFP		145		126			0.9:1
Lithium-ion batteries (LIBs)	U-KCMF(3:2)/rGO //LFP		145			339		2.3:1
	KCMG (3:2)/rGO //LFP		145				288	2.0:1
Lithium-ion capacitor/battery hybrids (LIC/Bs)	KCMF(3:2) //AC/LFP(1:3)			125	126			1:1
	U-KCMF(3:2)/rGO //AC/LFP(1:3)			125		339		2.7:1
	KCMF(3:2)/rGO //AC/LFP(1:3)			125			288	2.3:1

Table S7.Design of m_{\star}/m_{-} ratios for KCMF(3:2)//AC, U-KCMF(3:2)/rGO//AC,KCMF(3:2)/rGO//ACLICs, KCMF(3:2)//LFP, U-KCMF(3:2)/rGO//LFP,KCMF(3:2)/rGO//LFPLIBs and KCMF(3:2)//AC/LFP(1:3), U-KCMF(3:2)/rGO//AC/LFP(1:3), KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs.

Table S8. Performance summary of different energy storage devices in this study: KCMF(3:2)//AC, U-KCMF(3:2)/rGO//AC, KCMF(3:2)/rGO//AC LICs (a), KCMF(3:2)//LFP, U-KCMF(3:2)/rGO//LFP, KCMF(3:2)/rGO//LFP LIBs (b) and KCMF(3:2)//AC/LFP(1:3), U-KCMF(3:2)/rGO//AC/LFP(1:3), KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs (c).

	Working	Energy density/	Power density/	Cycling behavior/
LICs	voltage/V	Wh ka ⁻¹	kW ka ⁻¹	retention%, repeated
	voltage/ v	wii Kg	K W Kg	cycles, current density
		54 2~47 6	0.4~0.8	74%/1000/5A g ⁻¹
	0.40	12 2 2 2 1	1.6.2.1	70%/2000/5A g ⁻¹
	0~4.0	45.2~50.1	6.2 12.2	66%/3000/5A g ⁻¹
KCME(3,2)//AC		51.5~22.7	0.2~12.2	60%/5000/5A g ⁻¹
KUMF(5:2)//AU		75.0.66.4	0.4.0.9	70%/1000/5A g ⁻¹
	0.44	73.0~00.4 50.3- 52.0	1.7.3.4	59%/2000/5A g ⁻¹
	0~4.4	15 0 25 0	$1.7 \sim 3.4$	49%/3000/5A g ⁻¹
		43.0~33.0	0.8~13.7	37%/5000/5A g ⁻¹
	0~4.0	780 676	0204	83%/1000/5A g ⁻¹
		/8.0~0/.0	$0.2 \sim 0.4$	75%/2000/5A g ⁻¹
		38.0~49.7	0.8~1.5	67%/3000/5A g ⁻¹
U-KCMF(3:2)/rGO		38.1~25.9	3.0~6.0	43%/5000/5A g ⁻¹
//AC	0~4.4	00 4 75 7	0.2.0.4	34%/1000/5A g ⁻¹
		90.4~73.7	$0.2 \sim 0.4$	23%/2000/5A g ⁻¹
		03.0~32.8	$0.8 \sim 1.0$	18%/3000/5A g ⁻¹
		39.0~24.8	3.3~0.3	14%/5000/5A g ⁻¹
		711 (12	0.2.04	94%/1000/5A g ⁻¹
	0.40	/1.1~61.3	0.2~0.4	84%/2000/5A g ⁻¹
	0~4.0	52.6~43.6	0.8~1./	72%/3000/5A g ⁻¹
KCMF(3:2)/rGO		32.0~18.8	3.4~6.8	53%/5000/5A g ⁻¹
//AC		1150 977	0.2.05	83%/1000/5A g ⁻¹
	0~4.4	115.9~8/./	0.2~0.5	65%/2000/5A g ⁻¹
		/4.4~60.8	0.9~1.9	47%/3000/5A g ⁻¹
		44./~24.8	3./~/.4	29%/5000/5A g ⁻¹

(a)

LIBs	Working voltage/V	Energy density/ Wh kg ⁻¹	Power density/ kW kg ⁻¹	Cycling behavior/ retention%, repeated cycles, current density
KCMF(3:2)//LFP	0~4.7	209.7~177.5 151.1~123.7 96.7~69.0	0.9~1.7 3.5~6.8 13.2~24.3	72%/1000/5A g ⁻¹ 72%/2000/5A g ⁻¹ 72%/3000/5A g ⁻¹ 65%/5000/5A g ⁻¹
U-KCMF(3:2)/rGO //LFP	0~4.7	325.7~263.2 202.3~154.3 102.3~51.6	0.5~1.0 1.9~3.6 6.2~9.9	56%/1000/5A g ⁻¹ 51%/2000/5A g ⁻¹ 47%/3000/5A g ⁻¹ 43%/5000/5A g ⁻¹
KCMF(3:2)/rGO //LFP	0~4.7	307.6~266.1 210.4~158.0 112.5~68.6	0.6~1.1 2.1~3.9 7.2~12.4	72%/1000/5A g ⁻¹ 72%/2000/5A g ⁻¹ 64%/3000/5A g ⁻¹ 52%/5000/5A g ⁻¹

LIC/Bs	Working voltage/V	Energy density/ Wh kg ⁻¹	Power density/ kW kg ⁻¹	Cycling behavior/ retention%, repeated cycles, current density
KCMF(3:2)	0~4.4	147.6~121.0 100.5~78.5 57.0~28.0	0.7~1.5 2.9~5.4 10.0~16.0	66%/1000/5A g ⁻¹ 60%/2000/5A g ⁻¹ 53%/3000/5A g ⁻¹ 46%/5000/5A g ⁻¹
//AC/LFP(1:3)	0~4.7	150.0~123.9 103.6~86.4 69.8~51.1	0.7~1.4 2.8~5.6 10.9~20.0	45%/1000/5A g ⁻¹ 40%/2000/5A g ⁻¹ 33%/3000/5A g ⁻¹ 31%/5000/5A g ⁻¹
U-KCMF(3:2)/rGO //AC/LFP(1:3)	0~4.4	218.6~157.2 124.1~95.1 69.4~36.3	0.4~0.7 1.4~2.6 5.0~8.7	31%/1000/5A g ⁻¹ 23%/2000/5A g ⁻¹ 20%/3000/5A g ⁻¹ 17%/5000/5A g ⁻¹
	0~4.7	243.6~183.0 135.9~106.4 79.1~49.7	0.4~0.7 1.3~2.6 5.0~9.0	48%/1000/5A g ⁻¹ 45%/2000/5A g ⁻¹ 36%/3000/5A g ⁻¹ 24%/5000/5A g ⁻¹
KCMF(3:2)/rGO //AC/LFP(1:3)	0~4.4	229.1~187.8 159.0~131.1 104.7~78.7	0.4~0.9 1.7~3.3 6.3~12.2	71%/1000/5A g ⁻¹ 65%/2000/5A g ⁻¹ 56%/3000/5A g ⁻¹ 42%/5000/5A g ⁻¹
	0~4.7	234.7~190.1 149.5~113.6 85.4~57.5	0.4~0.9 1.7~3.2 6.0~10.9	68%/1000/5A g ⁻¹ 62%/2000/5A g ⁻¹ 54%/3000/5A g ⁻¹ 28%/5000/5A g ⁻¹

Table S9. A comparison	for some reported LI	is (a	a) and LIBs (b).
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LICs	Working voltage/V	Energy density/ Wh kg ⁻¹	Power density/ kW kg ⁻¹	Cycling behavior/ retention%, repeated cycles, current density	Refs	
Li ₃ VO ₄ /N-C//AC	1.0-4.0	136.4-24.4	0.532-11.02	87%/1500/2 A g ⁻¹	[3]. <i>Adv. Mater.</i> , 2017, 29 , 1700142	
TiNb2O7@C//CFs	0.8-3.2	110.4-20	0.1-5.46	77%/1500/0.2 A g ⁻¹	[4]. <i>Nano Energy</i> , 2015, 15 , 104-115	
TiO ₂ belt//Graphene	0-3.8	82-21	0.57-19	73%/600/1 A g ⁻¹	[5]. Small, 2015, 11 , 1470-147	
LiNi0.5Mn1.5O4//AC	1.5-3.25	19-8	0.13-3.5	81%/3000/1 A g ⁻¹	[6]. <i>Nano Energy</i> , 2015, 12 , 69-75	
TiO2@EEG//EEG	0.0-3.0	72-10	0.303-2.0	68%/1000/1.5 A g ⁻¹	[7]. Small, 2016, 12 , 6207-621	
	0.40	~4.0 71.1~18.8	0.2~6.8	94%/1000/5A g ⁻¹ *		
KCMF(3:2)/rGO	0~4.0			84%/2000/5A g ⁻¹ *		
// AC	0.44	115.9~24.8 0.2~7.4	83%/1000/5A g ⁻¹ *	_		
	0~4.4		0.2~7.4	65%/2000/5A g ⁻¹ *	_	
				71%/1000/5A g ⁻¹ *		
	0.44	220 1, 79 7	0 4 12 2	65%/2000/5A g ⁻¹ *	- This work	
KCMF(3:2)/rGO //AC/LFP(1:3)	0~4.4	229.1~/8./	0.4~12.2	88%/1000/5A g ⁻¹ #	T IIIS WOLK	
				81%/2000/5A g ⁻¹ #	_	
				68%/1000/5A g ⁻¹ *		
		2247 575	0.4.10.0	62%/2000/5A g ⁻¹ *	_	
	U~4./	234.1~31.3	0.4~10.9	88%/1000/5A g ⁻¹ #	-	
				81%/2000/5A g ⁻¹ #		

Note: * Based on the 1st cycle;

Based on the 50th cycle.

LIBs	Working voltage/V	Energy density/ Wh kg ⁻¹	Power density/ kW kg ⁻¹	Cycling behavior/ retention%, repeated cycles, current density	Refs
Si/graphene// LiNi _{1/3} Co _{1/3} Mn _{1/3} O ₂	3-4.3	156	0.03	70.4%/15/ 0.0375 A g ⁻¹	[8]. <i>Nano Energy</i> , 2012, 1 , 164-171
Li4Ti5O12-Li2Ti3O7//LFP	1.9-2.5	75	0.048	#	[9]. Adv. Funct. Mater., 2013, 23 , 640-647
Sn-C //Li[Ni0.45C00.1Mn1.45O4]	3.1-4.8	170	0.18	#	[10]. J. Am. Chem. Soc., 2011, 133 , 3139-3143
FeSb-TiC//LNMO	2.0-5.0	260	0.127	68%/50/ 0.0365 A g ⁻¹	[11]. Chem. Mater., 2014. 26 , 5905-5913
TiO2-MoO3//LiCoO2	1.0-4.0	285	1.086	#	[12]. Adv. Funct. Mater., 2015, 25 , 3524-3533
KCMF(3:2)/rGO//LFP	0~4.7	307.6~266.1 210.4~158.0 112.5~68.6	0.6~1.1 2.1~3.9 7.2~12.4	72%/2000/5A g ⁻¹ * 64%/3000/5A g ⁻¹ * 102%/2000/5A g ⁻¹ # 90%/3000/5A g ⁻¹ # 73%/5000/5A g ⁻¹ #	-
KCMF(3:2)/rGO //AC/LFP(1:3)	0~4.4	229.1~187.8 159.0~131.1 104.7~78.7 234.7~190.1	0.4~0.9 1.7~3.3 6.3~12.2 0.4~0.9	71%/1000/5A g ⁻¹ * 65%/2000/5A g ⁻¹ * 88%/1000/5A g ⁻¹ # 81%/2000/5A g ⁻¹ # 69%/30005A g ⁻¹ # 68%/1000/5A g ⁻¹ * 62%/2000/5A g ⁻¹ *	This work
	0~4.7	149.5~113.6 85.4~57.5	1.7~3.2 6.0~10.9	88%/1000/5A g ⁻¹ # 81%/2000/5A g ⁻¹ # 69%/3000/5A g ⁻¹ #	

Note: * Based on the 1st cycle;

Based on the 50th cycle.

Table S10. A comparison for advanced LICs (a), LIC/Bs (b) and LIBs (c) based on the recently reported perovskite fluorides anodes.

(a)					
LICs	Working voltage/V	Energy g density/ Wh kg ⁻¹	Power density/ kW kg ⁻¹	Cycling behavior/ retention%, repeated cycles, current density	Refs
KNCF(1:6)//AC	0~4.5	96~11	0.33~10.5	64%/1000/5 A g ⁻¹	[1]. J. Mater. Chem. A, 2019, 7, 8315-8326
NNCF(1:1)//AC	0~4.3	96.1~33.4	0.5~17.2	$68\%/1000/5 \mathrm{A g^{-1}}$	[2]. Chem. Commun., 2019, 55 , 6739-6742
KCMF(3:2)/rGO//AC	0~4.4	115.9~24.8	0.2~7.4	83%/1000/5A g ⁻¹	This work
(b)					
	Working	Energy	Power	Cycling behavior/	
LIC/Bs	voltage/V	density/	density/	retention%, repeated	Refs
	voltage/ v	Wh kg ⁻¹	kW kg ⁻¹	cycles, current density	
KNCF(1:6) //AC+LFP(1:1)	0~4.3	110~38	0.4~12.7	$73\%/1000/5 \text{ Ag}^{-1}$	[1]. J. Mater. Chem. A, 2019, 7, 8315-8326
NNCF(1:1) //AC+LFP(1:1)	0~4.6	132.3~53.1	0.6~18.4	$82\%/1000/5 \text{ Ag}^{-1}$	[2]. Chem. Commun., 2019, 55 , 6739-6742
KCMF(3:2)/rGO				71%/1000/5A g ⁻¹ (Based on the 1 st cycle)	
//AC/LFP(1:3)	0~4.4	229.1~78.7	0.4~12.2	88%/1000/5A g ⁻¹ (Based on the 50 th cycle)	This work
(c)					
LIBs	Workin voltage/	g density/ Wh kg ⁻¹	Power density/ kW kg ⁻¹	Cycling behavior/ retention%, repeated cycles, current density	Refs
KNCF(1:6)//LFP	0-4.7	173-29	0.75-15.3	67%/2000/2 A g ⁻¹	[1]. J. Mater. Chem. A, 2019, 7 , 8315-8326

NNCF(1:1)//LFP 1~4.7 196.2~35.2 0.8~19.2 KCMF(3:2)/rGO//LFP 0~4.7 307.6~68.6 0.6~12.4 — [2]. Chem. Commun.,

This work

2019, **55**, 6739-6742

63%/2000/3 A g⁻¹

72%/2000/5A g⁻¹ (Based on the 1st cycle)

102%/2000/5A g⁻¹ (Based on the 50th cycle)

Table S11. Performance summary of different energy storage devices in this study under high (40 $^{\circ}$ C) and low (-20 $^{\circ}$ C) temperature: KCMF(3:2)/rGO//AC LICs, KCMF(3:2)/rGO//LFP LIBs and KCMF(3:2)/rGO//AC/LFP(1:3) LIC/Bs.

					Cycling behavior/
Energy storage	т/0С	Working	Energy density/	Power density/	retention%,
devices	I/C	voltage/V	Wh kg ⁻¹	kW kg ⁻¹	repeated cycles,
					current density
			79.2~68.2	0.2~0.4	77%/250/2A g ⁻¹
	40		59.6~51.4	0.9~1.7	75%/500/2A g ⁻¹
KCMF(3·7)/#CO			42.9~30.6	3.4~6.9	65%/1000/2A g ⁻¹
		0~4.0V			82%/1000/2A g ⁻¹
//AC	20		53.0~28.6	0.2~0.4	77%/2000/2A g ⁻¹
	-20		19.0~9.1~1.9	0.9~1.7~3.4	75%/3000/2A g ⁻¹
					66%/5000/2A g ⁻¹
KCMF(3:2)/rGO //LFP			351.0~313.4	0.6~1.1	63%/250/2A g ⁻¹
	40		247.0~164.8	2.1~3.9	51%/500/2A g ⁻¹
			77.2~34.3	6.3~11.2	22%/1000/2A g ⁻¹
	20	0~4.7V			95%/1000/2A g ⁻¹
			115.7~65.4	0.5~1.0	93%/2000/2A g ⁻¹
	-20		37.3~21.3~8.7	1.9~3.2~5.7	90%/3000/2A g ⁻¹
					90%/5000/2A g ⁻¹
			262.5~221.4	0.4~0.8	77%/250/2A g ⁻¹
	40		189.3~146.5	1.6~3.1	66%/500/2A g ⁻¹
KCMF(3:2)/rGO			103.3~72.5	5.7~10.9	45%/1000/2A g ⁻¹
		0~4.4V			63%/1000/2A g ⁻¹
//AC/LFF(1:5)	20		41.0~25.1	0.3~0.7	61%/2000/2A g ⁻¹
	-20		17.3~9.6~4.6	1.4~2.6~5.5	58%/3000/2A g ⁻¹
					58%/5000/2A g ⁻¹

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