

Electronic Supplementary Information

An Iron-based Polyanionic Cathode for Potassium Storage with High Capacity and Excellent Cycling Stability

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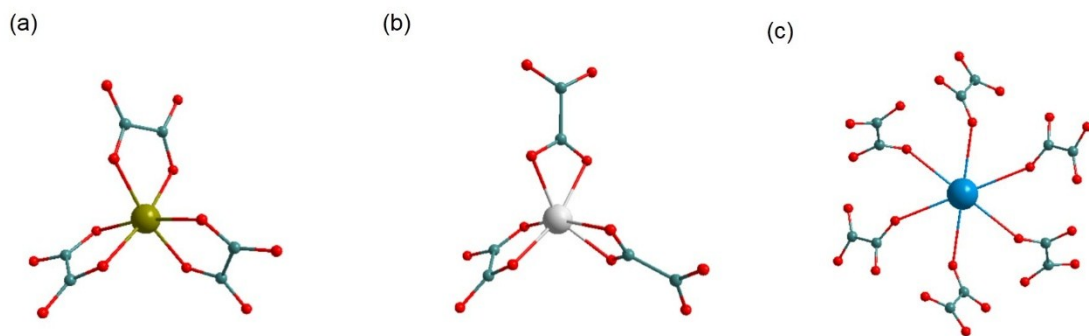


Fig. S1. Coordination of (a) Fe, (b) Li, and (c) K in KLFO.

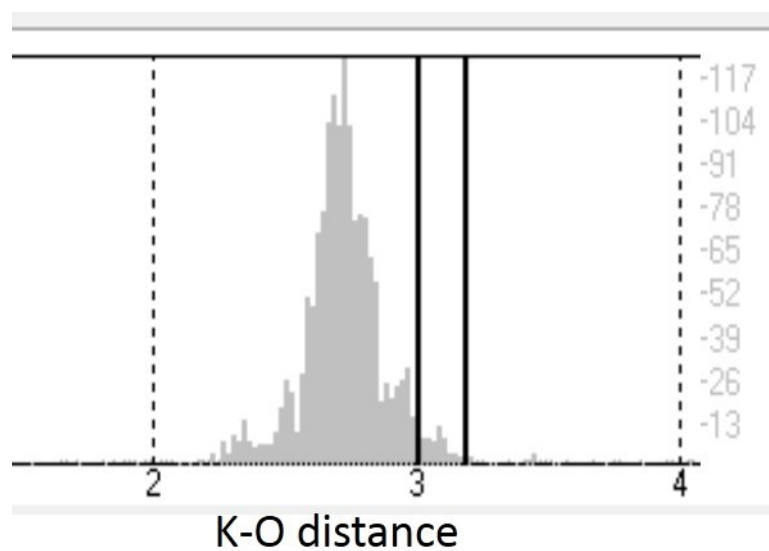


Fig. S2. K-O distance in KLFO in comparison with statistic values. Gray: 1681 compounds from statistic. Black: KLFO.

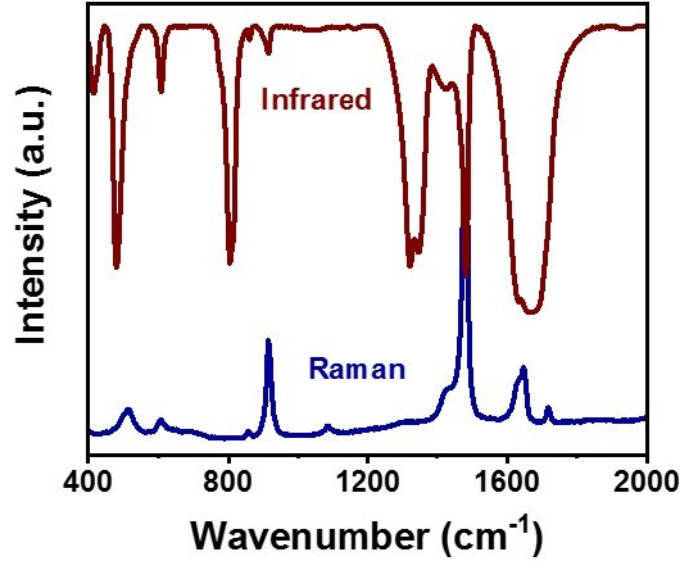


Fig. S3. FT-IR and Raman spectra of pristine KLFO sample.

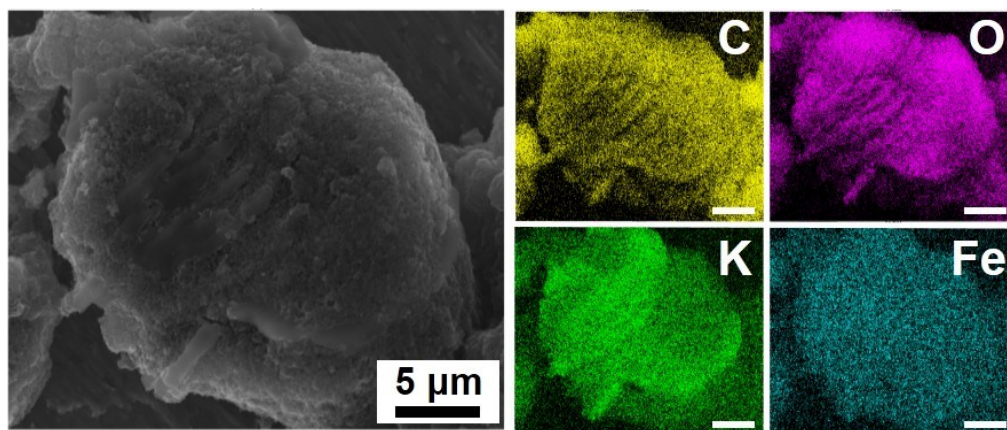


Fig. S4. SEM and EDS mapping of KLFO@C.

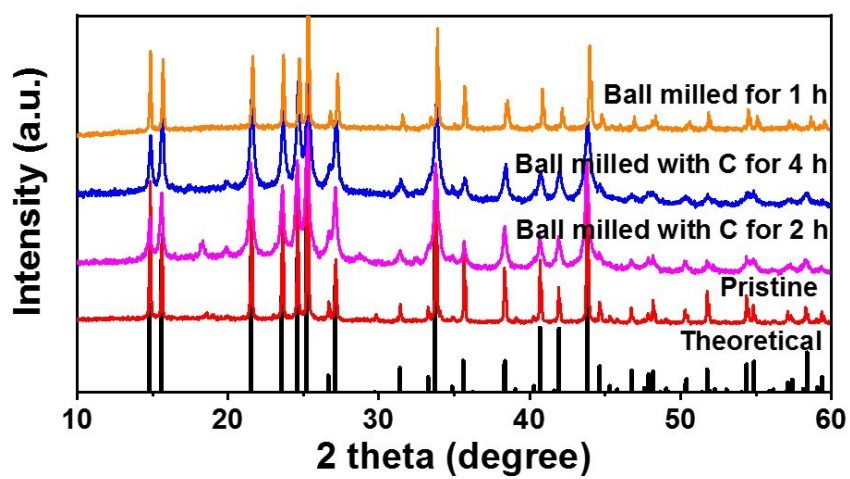


Fig. S5. Powder XRD patterns of samples at different stages.

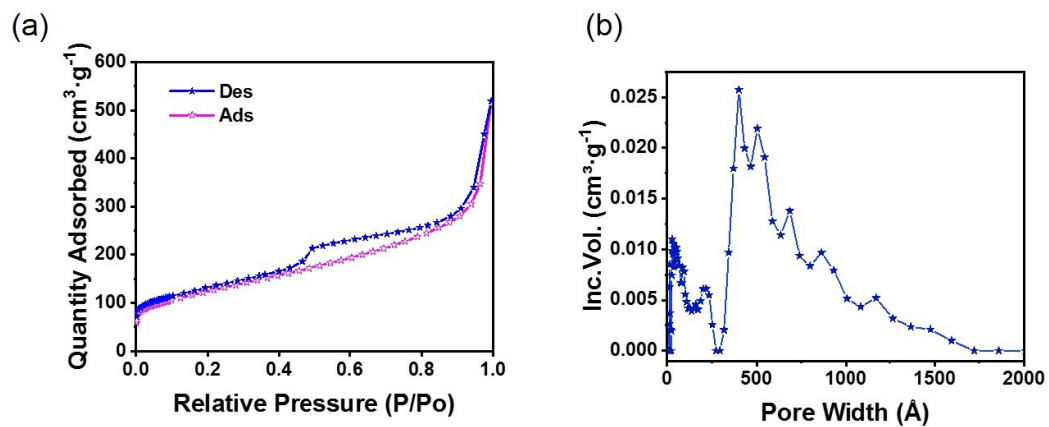


Fig. S6. Brunauer–Emmett–Teller surface area and pore structure of KLFO@C sample.

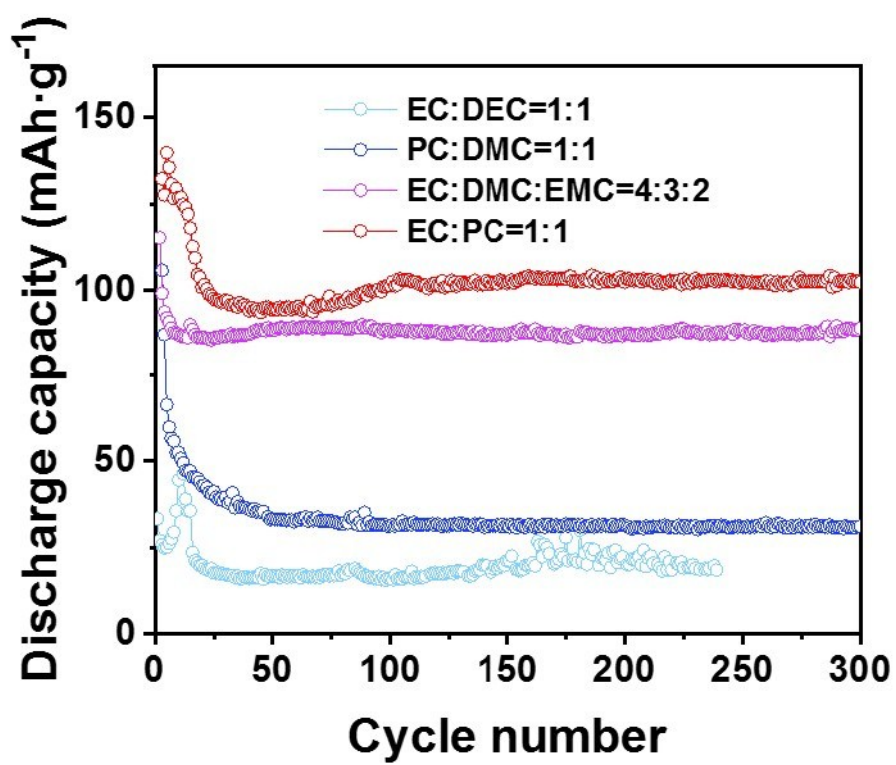


Fig. S7. Capacity retention of cells in preliminary electrolyte screening.

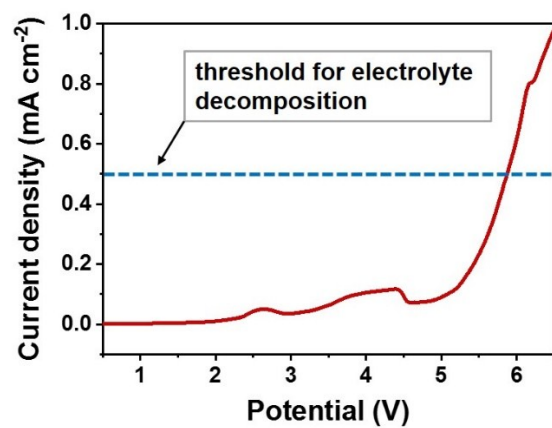


Fig. S8. Linear sweep voltammetry of the pure electrolyte cell.

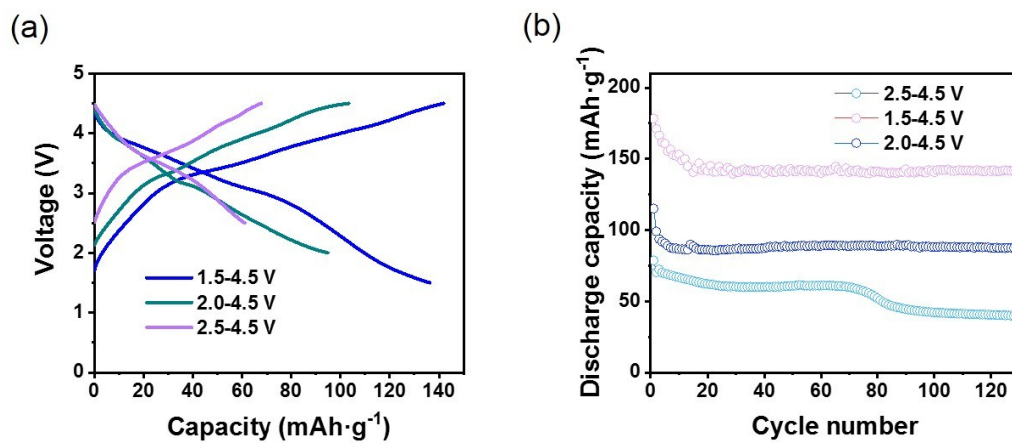


Fig. S9. Galvanostatic charging and discharging curves (a) and cycle stability (b) in different voltage windows of 2.5-4.5 V, 2.0-4.5 V and 1.5-4.5 V under the current density of 120 mA g^{-1} .

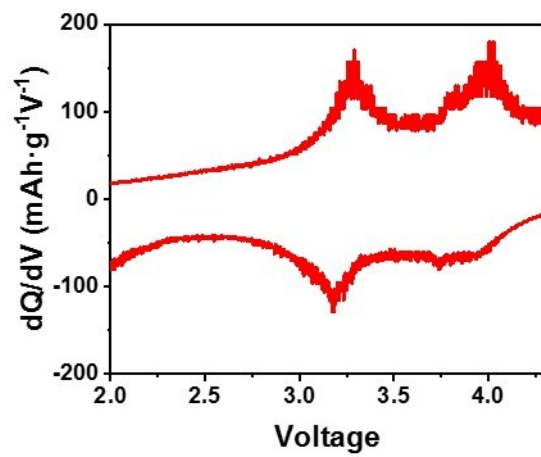


Fig. S10. dQ/dV curve of a cell cycled in 1.5-4.5 V under the current density of 120 mA g⁻¹, corresponding to GCD curve in Figure 2a.

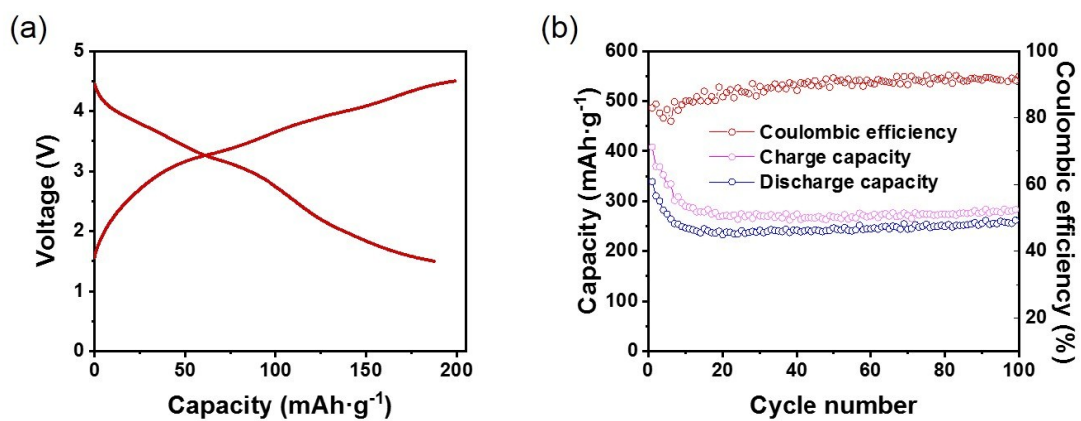


Fig. S11. The GCD curve and cycle stability of a cell under the current density of 25 mA g^{-1} .

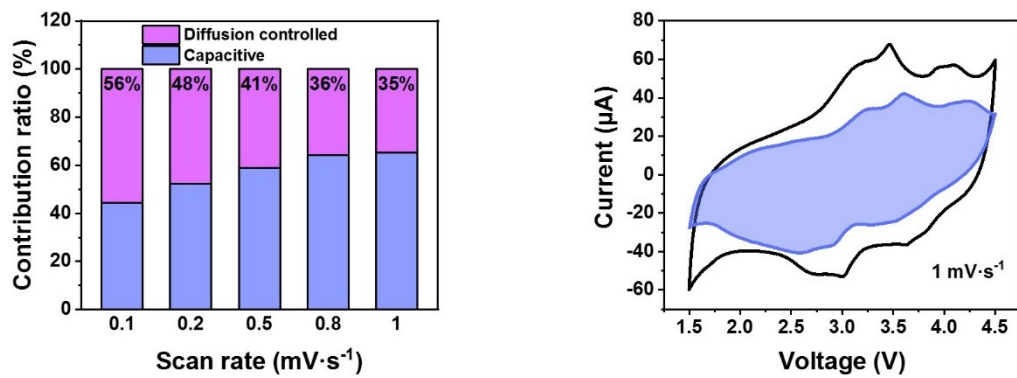


Fig. S12. Dynamic analysis. (a) Specific percentages of diffusion control at different scan rates. (b) Schematic illustration of capacitance-process (blue) at 1 mV s⁻¹.

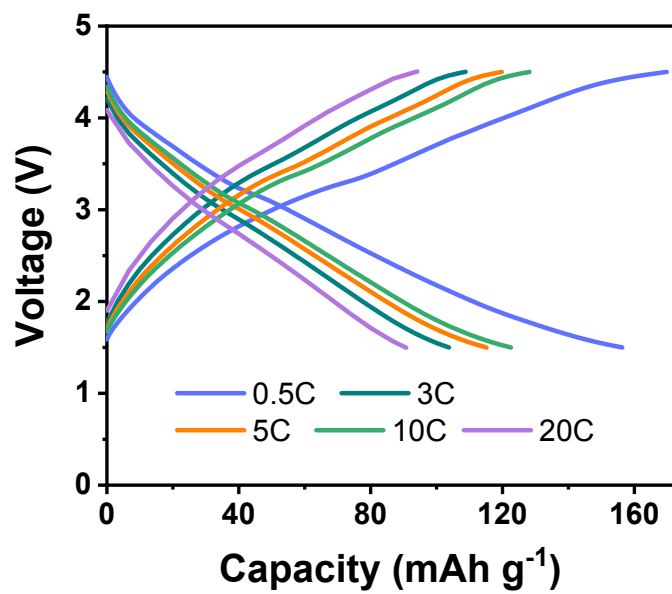


Fig. S13. GCD curves at different current densities. 1C = 120 mA g⁻¹.

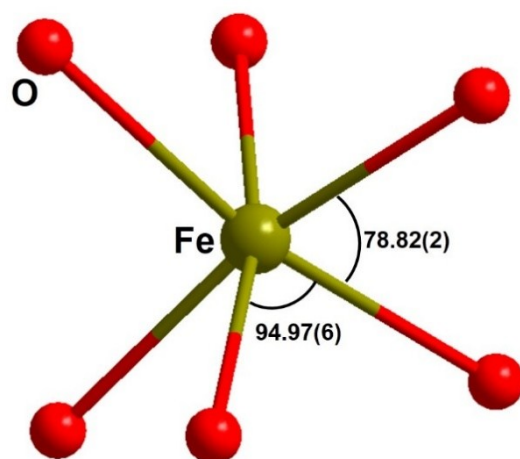


Fig. S14. Motif of [FeO₆] octahedron with maximum and minimum bond angles highlighted.

Table S1. Rietveld refinement of pristine $\text{KLi}_3\text{Fe}(\text{C}_2\text{O}_4)_3$.

Trigonal system, $R\bar{3}c$ (No. 167), Mr = 379.83 (g/mol), Z = 6

$a = 11.430(1) \text{ \AA}$, $b = 11.430(1) \text{ \AA}$, $c = 15.153(1) \text{ \AA}$, $V = 1714.5(4) \text{ \AA}^3$

$R_w = 0.0786$, $wR_p = 0.1027$, $\chi^2 = 1.535$

Atom	x/a	y/b	z/c	Multiplicity	U_{iso}
Fe	0	0	0.25	6	0.0288
K1	0.6667	0.3333	0.3333	6	0.0534
O1	0.3939	0.3072	0.3141	36	0.0394
O2	0.1851	0.1108	0.0.3193	36	0.0128
C1	0.2972	0.2149	0.2976	36	0.0115
Li1	0.2228	0.2228	0.25	18	0.0456

Table S2. Assignments of peaks in FTIR and Raman spectra of the pristine sample.

Peaks number	IR	Raman	Band assignments
1	1667(strong)		va (C=O)
2	1631(strong)		vs (C-O) + v(C-C)
3	1479(strong)		vs (C-O) + δ (O-C=O)
4	1425		vs (C-O) + δ (O-C=O)
5	1345(strong)		vs (C-O)
6	1319(strong)		δ (O-C=O)
7	912		vs (C-O) + δ (O-C=O)
8	860		δ (O-C=O)
9	804(strong)		v(M-O)
10	605		v(M-O) + v(C-C)
11	480 (strong)		Ring deform + δ (O-C=O)
i		1719	v (CO)
ii		1645(m)	v (CO)
iii		1625	v (CO)
iv		1481(s)	v (C=O) stretching
v		912(m)	v (C-C) stretching
vi		851	δ (O-C=O) bending
vii		601	δ (O-C=O) symmetric bending
viii		508(m)	v (MO ring)

Table S3. Fitting parameters of Fe K-edge EXAFS spectrum of pristine KLFO sample.

Path	Coordination Number	R	S_0^2	ΔE_0 (eV)	σ^2 (\AA^2)	<i>R factor</i>
Fe-O1	6	2.12(4)	0.71	2.982	0.01	0.019
Fe-C	6	2.87(4)	0.71	2.982	0.01	
Fe-O1-C	12	3.13(4)	0.71	2.982	0.01	
Fe-O2	6	4.08(11)	0.71	2.982	0.01	
Fe-C-O2	12	4.09(5)	0.71	2.982	0.01	

Table S4 Quantitative analysis of the oxidation state by linear combination fitting ofFe *K*-edge XANES.

Time	E (eV)	ΔE (eV)	oxidation state	%Fe ³⁺	%Fe ²⁺
Ref Fe ²⁺	7121.80	0.00	2.000	0	100
Ref Fe ³⁺	7126.80	5.00	3.000	100	0
20	7122.05	0.25	2.05	5	95
30	7122.42	0.62	2.124	12.4	87.6
40	7122.42	0.62	2.124	12.4	87.6
50	7122.49	0.69	2.138	13.8	86.2
60	7122.53	0.73	2.146	14.6	85.4
70	7122.61	0.81	2.162	16.2	83.8
80	7122.65	0.85	2.17	17	83
90	7122.9	1.1	2.22	22	78
100	7122.99	1.19	2.238	23.8	76.2
110	7123.72	1.92	2.384	38.4	61.6
120	7123.97	2.17	2.434	43.4	56.6
130	7124.44	2.64	2.528	52.8	47.2
140	7124.92	3.12	2.624	62.4	37.6
150	7125.37	3.57	2.714	71.4	28.6
160	7126.01	4.21	2.842	84.2	15.8
170	7126.75	4.95	2.99	99	1
180	7126.59	4.79	2.958	95.8	4.2
190	7125.83	4.03	2.806	80.6	19.4
200	7125.69	3.89	2.778	77.8	22.2
210	7125.07	3.27	2.654	65.4	34.6
220	7124.43	2.63	2.526	52.6	47.4
230	7124.18	2.38	2.476	47.6	52.4
240	7123.47	1.67	2.334	33.4	66.6
250	7123.05	1.25	2.25	25	75
260	7122.96	1.16	2.232	23.2	76.8
270	7122.63	0.83	2.166	16.6	83.4
280	7122.62	0.82	2.164	16.4	83.6
290	7122.54	0.74	2.148	14.8	85.2
300	7122.5	0.7	2.14	14	86
310	7122.44	0.64	2.128	12.8	87.2
320	7122.4	0.6	2.12	12	88
330	7122.31	0.51	2.102	10.2	89.8
340	7122	0.2	2.04	4	96

Table S5. Electrochemical performances of KLFO@C and cathode materials of recently reported for potassium-ion devices.

Cathode material	Type	Voltage [V]	Current density (mA g ⁻¹)	Stable capacity	Cycle number	Capacity after cycles (mA g ⁻¹)	Ref.
P2-type K _{2/3} Ni _{1/3} Co _{1/3} Te _{1/3} O ₂	oxide	1.25–5.0	6.5	30	25	30	6
P2-Type K _{0.63} Fe _{0.5} Mn _{0.5} O ₂	oxide	1.5–4.2	100	103	350	80	7
P2-type Na _{0.84-x} K _x CoO ₂	oxide	2.0–4.2	7.6	82	50	69	8
P2-type K ₂ Ni ₂ TeO ₆	oxide	1.3–4.7	6	70	70	62	9
P2-Type K _{0.6} CoO ₂	oxide	1.7–4.0	100	62	120	37	9
P2-type K _{0.6} CoO ₂	oxide	1.7–4.0	40	75	300	65	10
P3-type K _{0.45} MnO ₂	oxide	1.5–4.0	20	101	100	66.7	11
P2-type K _{0.3} MnO ₂	oxide	1.5–3.5	28	74	700	50	12
P2-type K _{0.41} CoO ₂	oxide	2.0–3.9	11.8	57	30	54	12
P3-type K _{2/3} CoO ₂	oxide	2.0–3.9	11.8	60	30	55	13
P3-K _{0.69} CrO ₂	oxide	1.5–3.8	100	100	1000	65	14
P3-K _{0.8} CrO ₂	oxide	1.5–3.9	10	90	300	50	15
P3-Type K _{0.5} MnO ₂	oxide	1.5–4.2	5	140	20	49	16
P3-Type K _{0.5} MnO ₂	oxide	1.5–3.9	5	110	20	86	17
P3-K _{0.54} [Co _{0.5} Mn _{0.5}]O ₂	oxide	1.5–4.0	500	78	500	48	18
P'3-type Na _{0.52} CrO ₂	oxide	2.0–3.6	500	52	200	51	19
O3-type KCrS ₂	oxide	1.8–3.0	9	71	1000	64	20
O3-type KCrO ₂	oxide	1.5–4.0	10	85	100	57	21
K _{0.37} Na _{0.3} Ni _{0.17} Co _{0.17} Mn _{0.66} O ₂	oxide	2.0–4.2	20	86.1	100	78.8	22
K _{0.67} Ni _{0.17} Co _{0.17} Mn _{0.66} O ₂	oxide	2.0–4.3	20	80	100	72	23
K _{0.7} Fe _{0.5} Mn _{0.5} O ₂	oxide	1.5–4.0	20	178	45	125	24
K _{1.06} Mn ₈ O ₁₆ /CNT	oxide	1.5–4.0	500	125	100	72.2	25
V ₂ O ₅ ·0.6H ₂ O	oxide	1.5–4.0	50	155	500	103	26
K _x V ₂ O ₅ ·nH ₂ O	oxide	2.0–4.3	20	226	50	167	27
K _{0.77} MnO ₂ ·0.23H ₂ O	oxide	1.5–4.0	1000	77	1000	62	28
K _{0.32} MnO ₂	oxide	2.0–4.5	100	49.2	100	36.1	29
Na _{0.9} Cr _{0.9} Ru _{0.1} O ₂	oxide	1.5–3.8	500	62	500	50.3	30
K _x MnO ₂ ·nH ₂ O	oxide	1.5–4.0	80	86	50	68	31
K _{0.45} Mn _{0.9} Mg _{0.1} O ₂	oxide	1.5–4.0	20	108	100	80.8	32
K _{0.28} MnO ₂ ·0.15H ₂ O	oxide	1.4–3.6	152	0.1C	100	123	33
AlF ₃ @ K _{1.39} Mn ₃ O ₆	oxide	1.5–4.0	10	110	100	105	34
K _{0.5} V ₂ O ₅	oxide	1.0–3.0	100	60	250	49	35
K ₂ V ₃ O ₈ /C	oxide	1.0–4.2	20	75	200	60	36
V ₂ O ₅	oxide	2.0–4.5	100	77.8	100	48	37
V ₂ O ₅ @rGO	oxide	1.5–4.3	147	222	500	178	38
V ₂ O ₅ @CNT	oxide	1.5–3.8	5	150	50	80	39
H ₂ V ₃ O ₈	oxide		5	168	100	126	40
K _{0.486} V ₂ O ₅	oxide	1.5–4.2	20	159	100	44.3	41
KFeO ₂	oxide	1.7–3.7	10	60	50	30	42
KMn _{7/6} Co _{0.4} O ₁₆	oxide	1.5–4.0	100	174	50	75	43
KNi _{1/3} Mn _{2/3} O ₂	oxide	1.5–4.5	86	82	200	73	44
K _{0.3} Ti _{0.75} Fe _{0.25} [Fe(CN) ₆] _{0.95} ·2.8H ₂ O	PBAs	1.0–4.5	100	113	100	73.1	45
K _{0.220} Fe[Fe(CN) ₆] _{0.805} ·4.01H ₂ O	PBAs	2.0–4.0	50	74.5	50	73.2	46
K _{1.6} Mn[Fe(CN) ₆] _{0.96} ·0.27H ₂ O	PBAs	3.2–4.3	50	110	30	86	47
K _{1.70} Mn[Fe(CN) ₆] _{0.90} ·1.10H ₂ O	PBAs	2.5–4.6	156	110	100	100	48
K _{1.75} Mn[Fe(CN) ₆] _{0.93} ·0.16H ₂ O	PBAs	2.0–4.5	30	120	100	116	47
K _{1.89} Mn[Fe(CN) ₆] _{0.92} ·0.75H ₂ O	PBAs	2.5–4.6	0.2C	142.4			49
K _{1.64} Fe[Fe(CN) ₆] _{0.89} ·0.15H ₂ O	PBAs	2.0–4.5	30	122	100	110	50
K _{1.88} Zn _{2.88} [Fe(CN) ₆] ₂ (H ₂ O) ₅	PBAs	3.4–4.15	14	55.6	100	52.8	51

$K_{1.92}Fe[Fe(CN)_6]_{0.94} \cdot 0.5H_2O$	PBA _s	2.0–4.3	13	133	200	123	52
$K_{1.7}Fe[Fe(CN)_6]_{0.9}$	PBA _s	2.0–4.5	100	120	300	78	53
$K_{1.59}Fe_{2.20}(CN)_6$	PBA _s	2.25–4.25	77.5	148	1000	99	53
$K_{1.68}Fe_{1.09}[Fe(CN)_6] \cdot 2.1H_2O$	PBA _s	2.0–4.5	20	110	100	81	53
$K_{1.55}Co_{0.88}[Fe(CN)_6] \cdot 3.2H_2O$	PBA _s	2.0–4.5	20	60	15	38.4	53
$K_{1.51}Ni_{1.05}[Fe(CN)_6] \cdot 2.1H_2O$	PBA _s	2.0–4.5	20	63	15	58.6	54
$K_{1.40}Cu_{0.93}[Fe(CN)_6] \cdot 2.1H_2O$	PBA _s	2.0–4.5	20	36	15	30	55
$KFe[Fe(CN)_6]$	PBA _s	2.0–4.5	100	90.7	1000	73	56
$KFe[Fe(CN)_6]$	PBA _s	0.8–2.1	2000	72	50	58	57
$KFe^{3+}[Fe^{2+}(CN)_6]$	PBA _s	2.5–4.2	8.7	78	500	69	58
$K_{1.85}Mn[Fe(CN)_6]_{0.98} \cdot 0.7H_2O$	PBA _s	2.0–4.5	15	126	120	120.1	59
$K_{1.81}Mn[Fe(CN)_6]_{0.97} \cdot 0.086H_2O$	PBA _s PBA _s	2.0–4.5	10	57	1000	50	60
$K_{1.4}Fe_4[Fe(CN)_6]_3$	PBA _s	2.0–4.0	50	72	40	57.6	61
$K_{1.62}Fe[Fe(CN)_6]_{0.92} \cdot 0.33H_2O$	PBA _s	2.5–4.5	50	120.9	100	117	62
5% Ni-doped PB	PBA _s	2.0–4.2	100	135	300	112	63
RGO@PB@SSM	PBA _s	2.0–4.0	50	61.4	305	46	64
KHCF@PPy	PBA _s	2.0–4.2	50	88.8	500	77.1	65
$K_2FeFe(CN)_6$	PBA _s	2.0–4.3	20	110	100	89	66
$FeFe(CN)_6$	PBA _s	0.0–0.975	111	140	20	119	67
$K_4Fe(CN)_6$	PBA _s	2.0–3.8	20	65.5	400	48.8	68
$FeFe(CN)_6$	PBA _s	1.5–4.0	625	100	500	93	69
<i>o</i> -Na ₂ C ₆ H ₂ O ₆	Organic	1.0–3.0	25	98.8	100	65.1	70
<i>p</i> -Na ₂ C ₆ H ₂ O ₆	Organic	1.0–3.0	25	228.5	50	190	71
CuTCNQ	Organic	2.0–4.0	50	206	50	170	72
HAT-containing polymer	Organic	0.9–3.4	10000	150	4500	150	73
PTCDA	Organic	1.5–3.5	50	117	200	90	74
PTCDA	Organic	1.2–3.2	10	87	300	63	75
PTCDA	Organic	1.5–3.5	1000	113	1000	98	76
AQDS	Organic	1.4–3.0	13	95	100	78	77
AQDS	Organic	1.4–3.0	390	80	1000	64	78
PAQS	Organic	1.5–3.4	200	106	200	68	79
PTPAn	Organic	2.0–4.0	100	75	60	71	80
PVK	Organic	2.0–4.7	500	104	400	73	81
PAN	Organic	2.0–4.0	50	100	100	98	82
SR	Organic	1.5–3.8	125	118	100	82	83
PQ-1, 5	Organic	1.2–3.2 V	250	115	200	105	84
PI-CMP	Organic	1.5–3.5 V	1000	109	1000	80.8	85
PPTS	Organic	0.8–3.2	100	282.3	-	-	86
<i>p</i> -DPPZ	Organic	2.5–4.5	162	200	1000	120	87
Na ₂ AQ ₂₆ DS	Organic	1.5–3.7	100	105	200	105	88
OHTAP	Organic	1.1–2.8	0.1C	220	50	162	89
Polydiaminoanthraquinone	Organic	1.2–3.2	250	120	200	105	90
PI@G	Organic	1.4–3.5	100	142	500	118	91
PTCDI	Organic	1.4–3.8	100	157	600	120	92
$K_2[(VO)_2(HPO_4)_2(C_2O_4)]$	Polyanion	2.0–4.6	22	65	200	54	93
$VOPO_4 \cdot 2H_2O$	Polyanion	-0.2–1.3	27	88.3	100	76	93
$FePO_4$	Polyanion	1.5–3.5	5	156	50	134	94
$K_3V_2(PO_4)_3/C$	Polyanion	2.5–4.3	20	55	100	52	95
$KTi_2(PO_4)_3/C$	Polyanion	1.2–2.8	64	75.6	100	82	96
$KVPO_4F$	Polyanion	2.0–4.8	6.65	72	50	65	96
$KVOPO_4$	Polyanion	2.0–4.8	6.65	73	50	69	96
$KVOPO_4$	Polyanion		24	115	100	60	97
$KVPO_4F$	Polyanion	3.0–5.0	5	100	30	78	98
KVP_2O_7	Polyanion	2.0–5.0	25	60	100	51	99
$KMoP_2O_7$	Polyanion	2.0–5.0	1C	25	-	-	100
$KTiP_2O_7$	Polyanion	2.0–5.0	1C	22	-	-	101
$K_3V_2(PO_4)_3$	Polyanion	0.01–3.0	25	88	500	78	102
$K_3V_2(PO_4)_2F_3$	Polyanion	2.0–4.5	10	101	100	98	103
$K(Mn,Co)F_3$	Others	1.2–4.2	40	132.6	60	103.4	104
I_2	Others	1.9–3.5	100	115	500	82	105
KLFO@C	Polyanion	1.5–4.5	120	140	5000	130	This

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