Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.
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Supplemnetray Information

New P2-type layered oxide cathode with superior full-cell performances for K-ion battery

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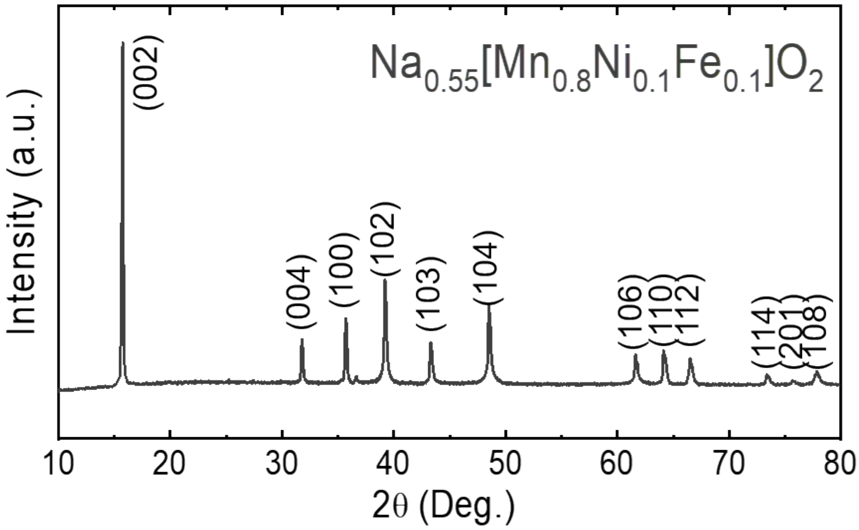


Fig. S1 XRD pattern of the $Na_{0.55}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$ powder.

Table S1. Chemical composition of $Na_{0.55}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$ confirmed by inductively coupled plasma analysis.

	Metal stoichiometry determined by inductively-coupled plasma optical emission spectrometry (ICP-OES)			
Formula	Na	Mn	Ni	Fe
$Na_{0.55}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$	0.549	0.795	0.102	0.103

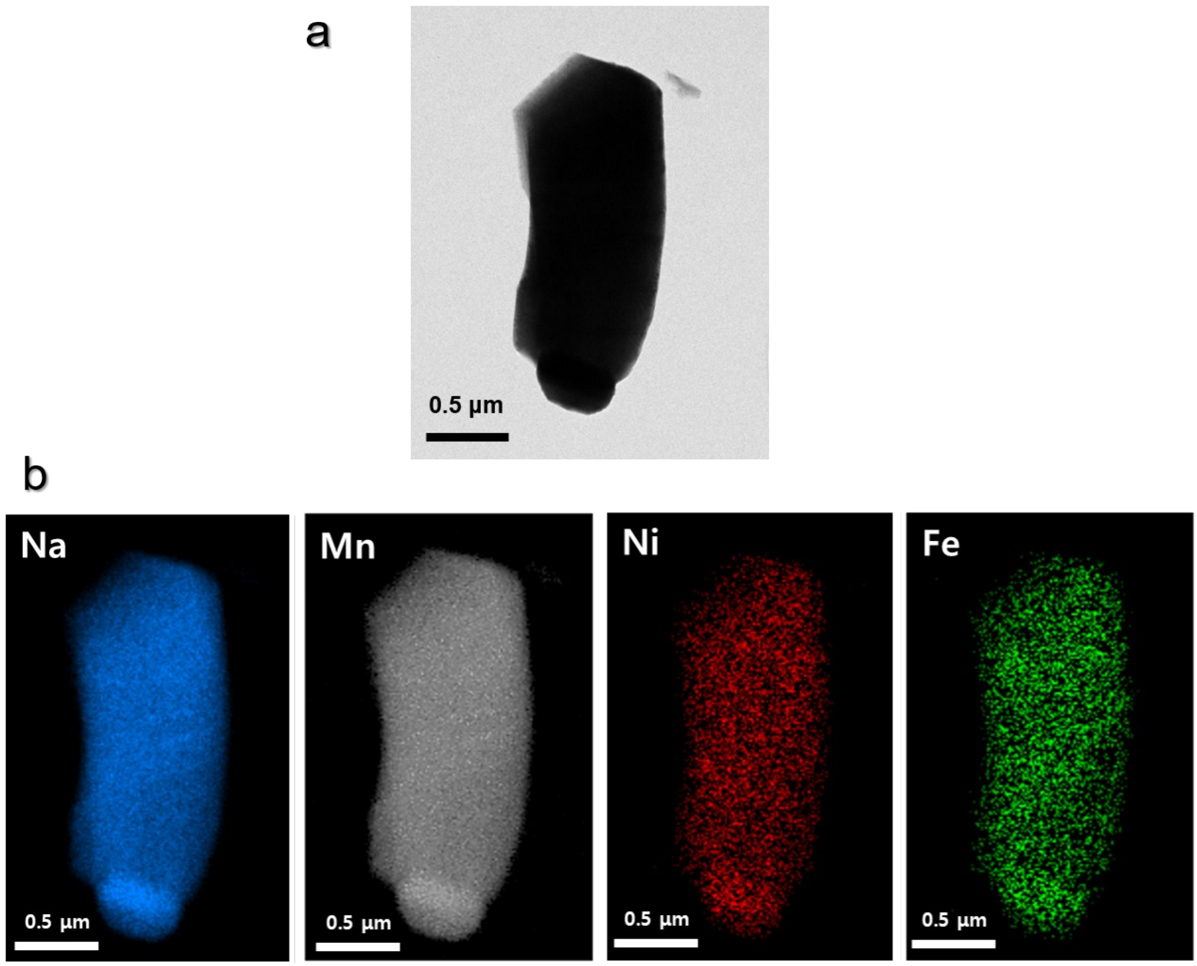


Fig. S2 (a) TEM image and (b) corresponding EDX maps of $Na_{0.55}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$.

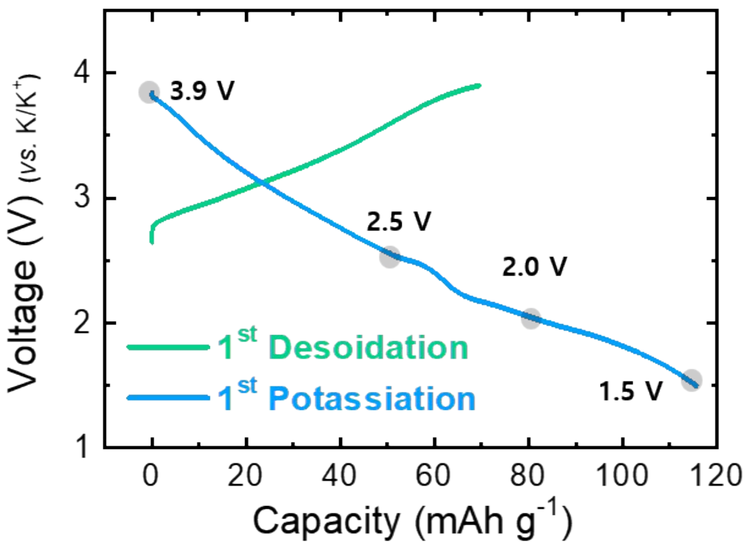


Fig. S3 Electrochemical ion exchange process in first cycle of $Na_xK_yMNFO_2$ electrode between 1.5 and 3.9 V.

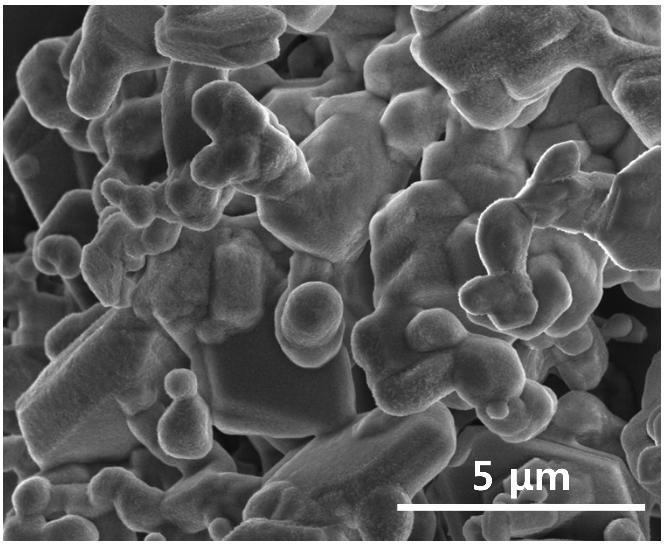


Fig. S4 SEM image of P2-$K_{0.75}MNFO_2$.

Table S2. Chemical composition of $K_{0.75}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$ confirmed by ICP analysis.

Metal stoichiometry determined by ICP-OES				
Formula	K	Mn	Ni	Fe
$K_{0.75}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$	0.748	0.796	0.103	0.101

Table S3. Detailed structural information of P2-$K_{0.75}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$ calculated by Rietveld refinement.

Atom	**Multiplicity**	***x***	***y***	***z***	B_{iso}	**Occupancy**
K1	2*b*	0	0	0.25	1.2(7)	0.31
K2	2*d*	0.66667	0.33333	0.25	0.7(7)	0.44
Mn1	2*a*	0	0	0	0.22(18)	0.8
Fe2	2*a*	0	0	0	0.22(18)	0.1
Ni1	2*a*	0	0	0	0.22(18)	0.1
O1	4*f*	0.33333	0.66667	0.0854(2)	1.0(5)	1

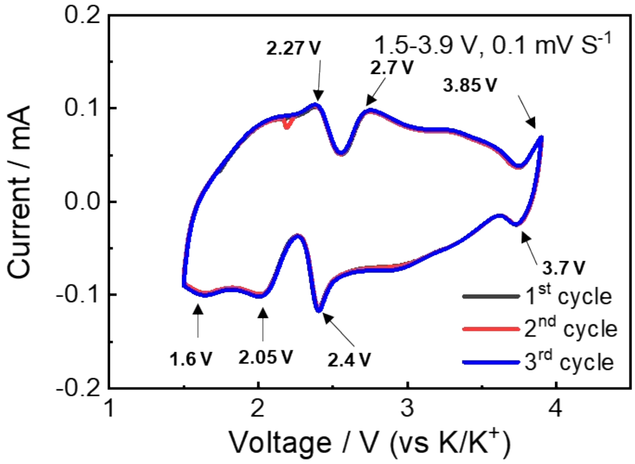


Fig. S5 CV curve of the $K_{0.75}MNFO_2$ cathode.

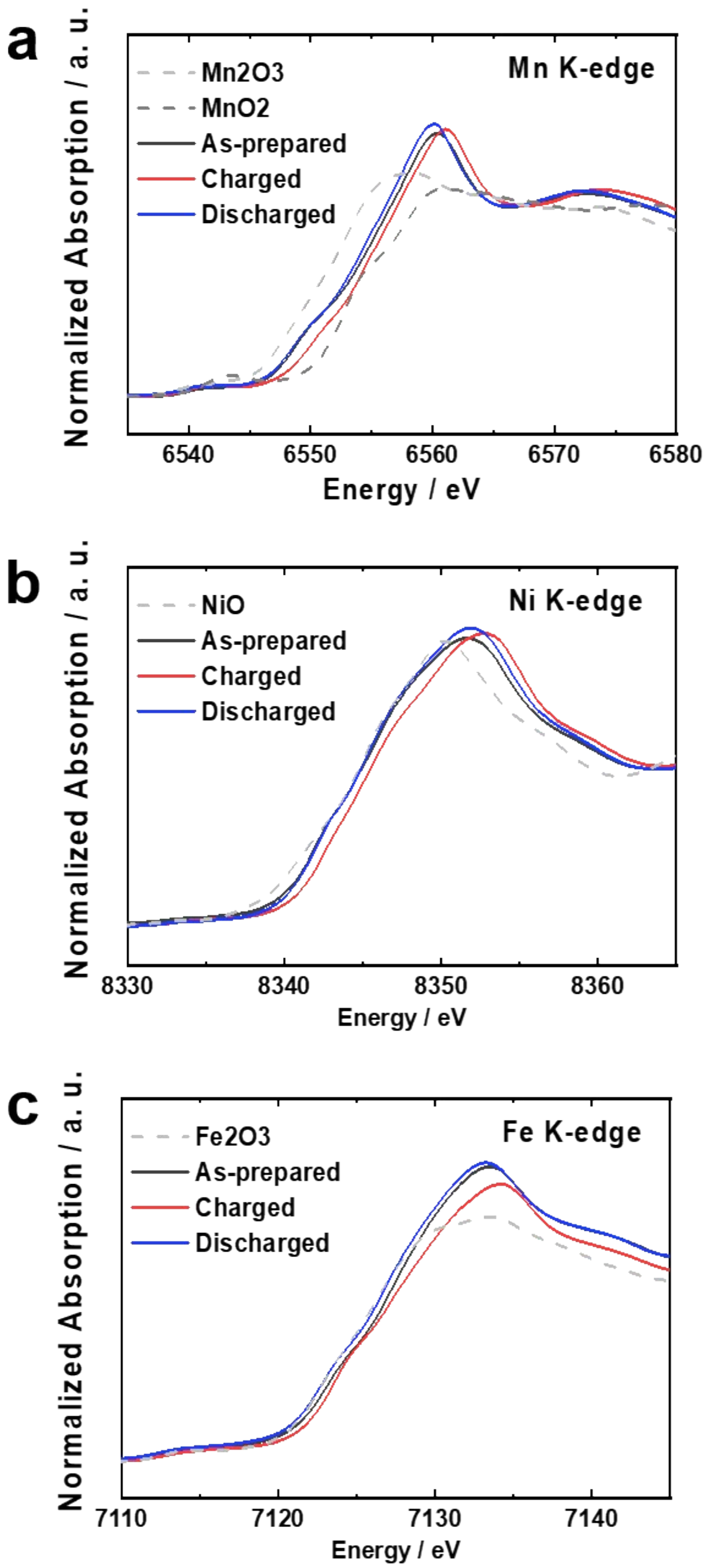


Fig. S6 XANES sepctra for $K_{0.75}MNFO_2$ measured at the end of charge (3.9 V) and discharge (1.5 V): (a) Mn, (b) Ni, and (c) Fe.

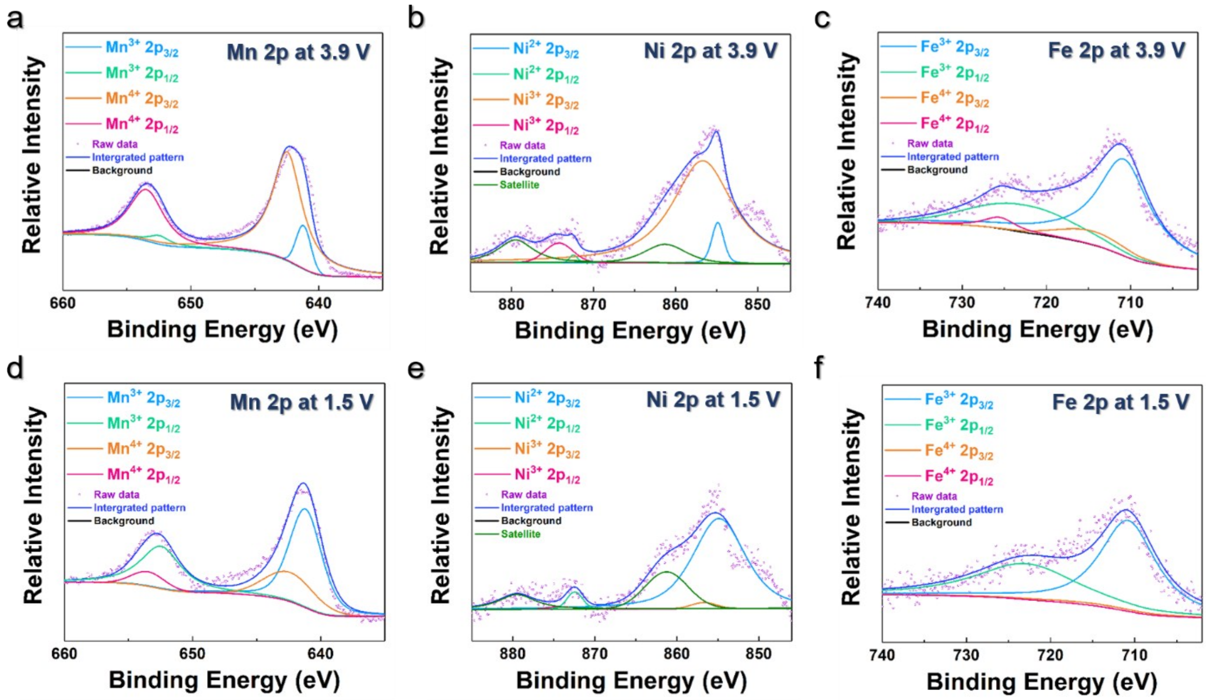
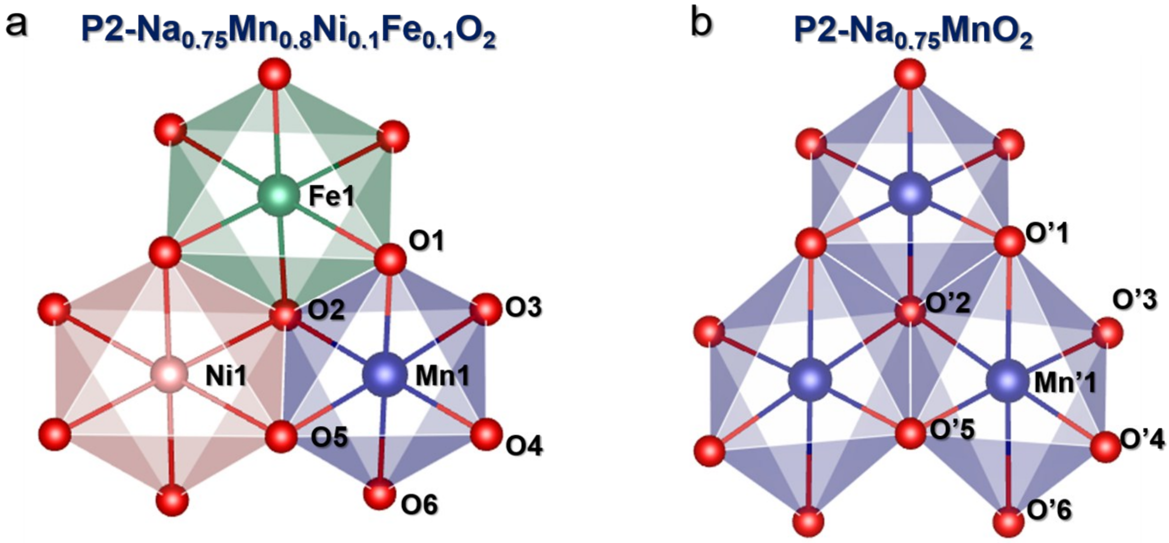


Fig. S7 XPS spectra of the $K_{0.75}MNFO_2$ cathode at charge end and discharge end states. Mn 2p spectra at (a) 3.9 V and (d) 1.5 V. Ni 2p spectra at (b) 3.9 V and (e) 1.5 V. Fe 2p spectra at (c) 3.9 V and (f) 1.5 V.



Bonding	Length (Å)
Mn1-O1	1.95257
Mn1-O2	1.95617
Mn1-O3	1.99616
Mn1-O4	1.94726
Mn1-O5	1.98666
Mn1-O6	1.99814

Bonding	Length (Å)
Mn'1-O'1	2.34663
Mn'1-O'2	1.99550
Mn'1-O'3	1.95339
Mn'1-O'4	2.03048
Mn'1-O'5	1.94072
Mn'1-O'6	2.34480

Fig. S8 Predicted Mn–O bond lengths in (a) P2-K_xMNFO_2 and (b) P2-K_xMnO_2, for comparison of the effect of Jahn–Teller distortion caused by the Mn^{3+} ions.

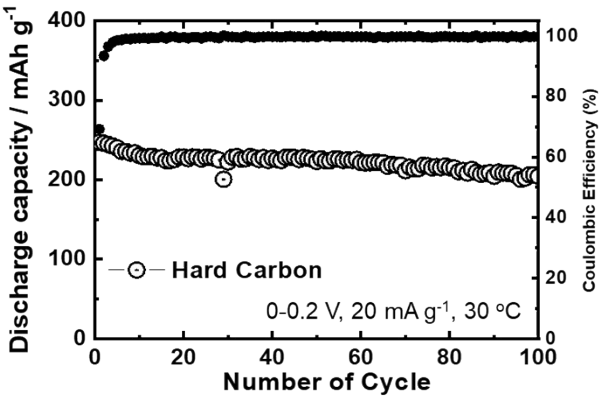


Fig. S9 Cycling stability of hard carbon anode in K-ion cell.

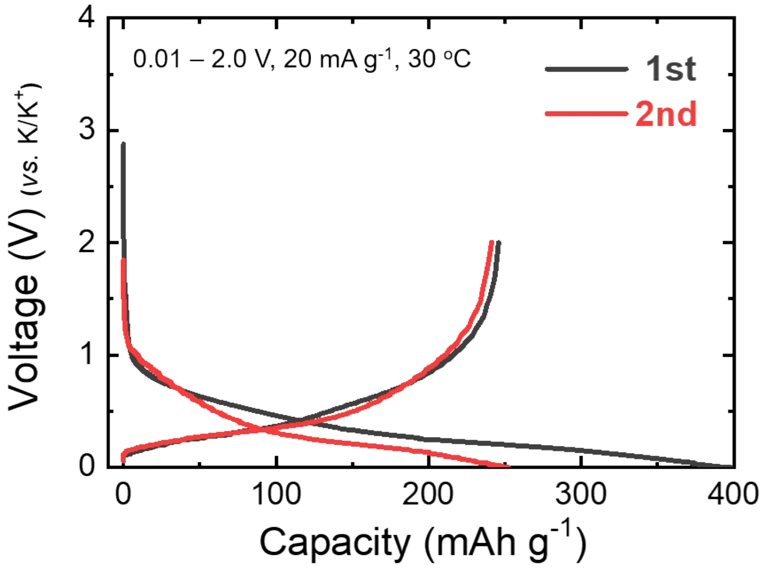


Fig. S10 Charge–discharge curves of hard carbon electrode in the voltage range of 0.01–2.0 V at 20 mA g^{-1}.

Table S4. Comparison of K storage performances of $K_{0.75}[Fe_{0.1}Ni_{0.1}Mn_{0.8}]O_2$ with previously reported layered oxide cathodes.

Cathode	Cut-off Potential	1st Discharge Capacity	Cycling Performance	Rate Performance	Full Cell	Ref
P2- $K_{0.41}CoO_2$	2.0-3.9	57 mAh/g at 11.8 mA/g	96% (30cycles) at 11.8 mA/g	66% at 472 mA/g	X	S1
P3- $K_{2/3}CoO_2$	2.0-3.9	60 mAh/g at 11.8 mA/g	96% (30cycles) at 11.8 mA/g	-	X	S1
P2- $K_{0.6}CoO_2$	1.7-4.0	80 mAg/g at 2 mA/g	60% (120cycles) at 100 mA/g	55% at 150 mA/g	O	S2
P2- $K_{0.6}CoO_2$	1.7-4.0	74 mAh/g at 40 mA/g	87% (300cycles) at 40 mA/g	50% at 200 mA/g	O	S3
P2- $Na_{0.84-x}K_xCoO_2$	2.0-4.2	82 mAh/g at C/20	85% (50cycles) at C/10	65% at 1C	X	S4
P'2-$K_{0.3}MnO_2$	1.5-3.5	74 mAh/g	68% (685cycles) at 27.9 mA/g	54%	O	S5
P3- $K_{0.5}MnO_2$	1.5-3.9	140 mAh/g	70% (50cylces) at 20 mA/g	-	X	S6
P3- $K_{0.45}MnO_2$	1.5-4.0	128.6 at 20 mA/g	70.8% (100cycles) at 20 mA/g	40% at 200 mA/g	X	S7
$K_{0.67}Ni_{0.17}Co_{0.17}Mn_{0.66}O_2$	2.0-4.3	76.5 mAh/g at 20 mA/g	87% (100cycles) at 20 mA/g	71% at 100 mA/g	X	S8
$K_{0.37}Na_{0.3}Ni_{0.17}Co_{0.17}Mn_{0.66}O_2$	2.0-4.2	86.1 mAh/g at 20 mA/g	91.5% (100cycles) at 20 mA/g	79% at 100 mA/g	X	S9
$K_{0.7}Fe_{0.5}Mn_{0.5}O_2$ nanowire	1.5-4.0	178 mAh/g at 20 mA/g	87% (450cycles) at 1000 mA/g	38% at 1000 mA/g	O	S10
P2-$K_{0.65}Fe_{0.5}Mn_{0.5}O_2$ microsphere	1.5-4.2	151 mAh/g at 20mA/g	60% (350cycles) at 100 mA/g	23% at 800 mA/g	O	S11
P'3- $K_{0.52}CrO_2$	2.0-3.4	89 mAh/g at 12.5 mA/g	73% (200cycles) at 500 mA/g	56% at 1250 mA/g	X	S12
P'3- $K_{0.8}CrO_2$	1.5-3.9	90 mAh/g at 10.9 mA/g	94% (300cycles) at 218 mA/g	57% at 436 mA/g	X	S13

P3- $K_{0.69}CrO_2$	1.5-3.8	100 mAh/g at 10 mA/g	65% (1000cycles) at 100 mA/g	65% at 1000 mA/g	X	S14
O3- $KCrO_2$	1.5-4.0	92 mAh/g at 5 mA/g	67% (100cycles) at 5 mA/g	33% at 500 mA/g	X	S15
O3- $KCrS_2$	1.8-3.0	74 mAh/g at 8.65 mA/g	90% (1000cycles) at 173 mA/g	58% at 865 mA/g	X	S16
P3- $K_{0.54}[Co_{0.5}Mn_{0.5}]O_2$	1.5-3.9	120 mAh/g	85% (500cycles) at 500 mA/g	65% at 500 mA/g	O	S17
P3- $K_{0.5}[Ni_{0.1}Mn_{0.9}]O_2$	1.5-3.9	121 mAh/g at 10 mA/g	82% (100cycles) at 10 mA/g	62% at 500 mA/g	X	S18
P3- $K_{0.45}Mn_{0.5}Co_{0.5}O_2$	1.5-3.9 1.2-3.9	89 mAh/g 140 mAh/g at 10 mA/g	80% (50cycles) at 50 mA/g (1.2-3.9V)	49% at 100 mA/g (1.2-3.9V)	X	S19
P3- $K_{0.45}Mn_{0.8}Fe_{0.2}O_2$	1.5-4.0	106.2 mAh/g at 20 mA/g	45% (100cycles) at 200 mA/g	61% at 200 mA/g	X	S20
P2-$K_{0.75}[Mn0._{8}Ni_{0.1}Fe_{0.1}]O_2$	1.5-3.0 V	110 mAh/g at 10 mA/g	70 % (200 cycles) at 1000 mA/g	57 % at 1000 mA/g	O	This work

Table S5. Comparison of the full cell performances of P2-$K_{0.75}[Mn_{0.8}Ni_{0.1}Fe_{0.1}]O_2$/hard carbon full cell with previously reported K-ion full cells using layered oxide cathode and carbonaceous anode.

Cathode	Cut-off Potential	1st Discharge Capacity	Cycling Performance	Ref
P2- $K_{0.6}CoO_2$ // Graphite	0.5-3.9	53 mAh/g at 3 mA/g	50% (5cycles) at 3 mA/g	S2
P2- $K_{0.6}CoO_2$ // Hard carbon	0.5-3.8	72 mAh/g at 30 mA/g	86% (50cycles) at 30 mA/g	S3
P'2- $K_{0.3}MnO_2$ // Hard carbon + Carbon black	0.5-3.4	90 mAh/g at 32 mA/g	76% (100cycles) at 32 mA/g (1.5-3.5V)	S5
$K_{0.7}Fe_{0.5}Mn_{0.5}O_2$ nanowire // Soft carbon	0.5-3.5	119 mAh/g	76% (250cycles)	S10

		at 20 mA/g	at 100 mA/g	
$K_{0.65}Fe_{0.5}Mn_{0.5}O_2$ microsphere	0.5-3.5	76 mAh/g at 100 mA/g	80% (100cycles) at 100 mA/g	S15
P3- $K_{0.54}[Co_{0.5}Mn_{0.5}]O_2$	0.5-3.6	95 mAh/g at 20 mA/g	82% (100cycles) at 20 mA/g	S17
P2-$K_{0.75}[Mn0_{.8}Ni_{0.1}Fe_{0.1}]O_2$	0.5-3.5	60 mAh/g at 20 mA/g	60 % (1000 cycles) at 20 mA/g	This Work

References in Table S4 and R5.

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