Supplementary Information (SI) for Materials Chemistry Frontiers.

Supporting information

Food-Additive-Inspired Bifunctional Proton Buffering-Iron Capture Route to Low-Defect Prussian Blue Cathodes for Sodium-Ion Batteries

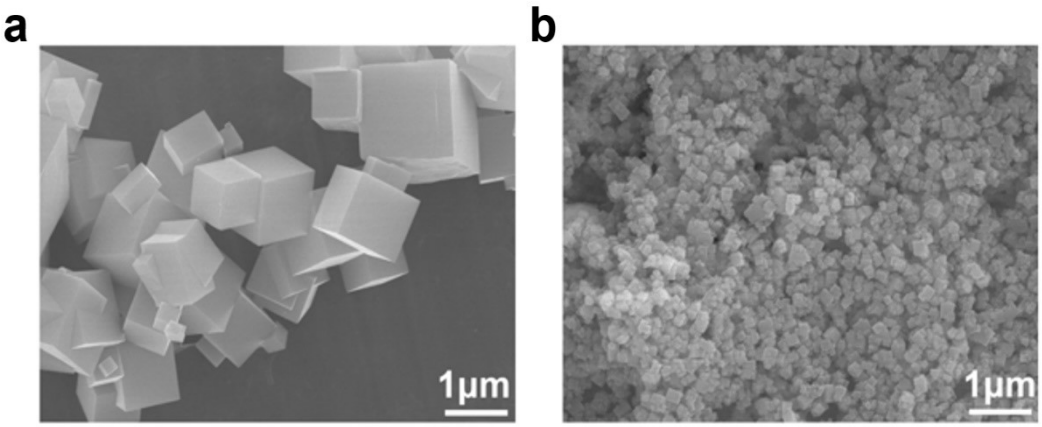

Fig. S1 SEM images of (a) PB, and (b) $InsP_6$-PB-30.

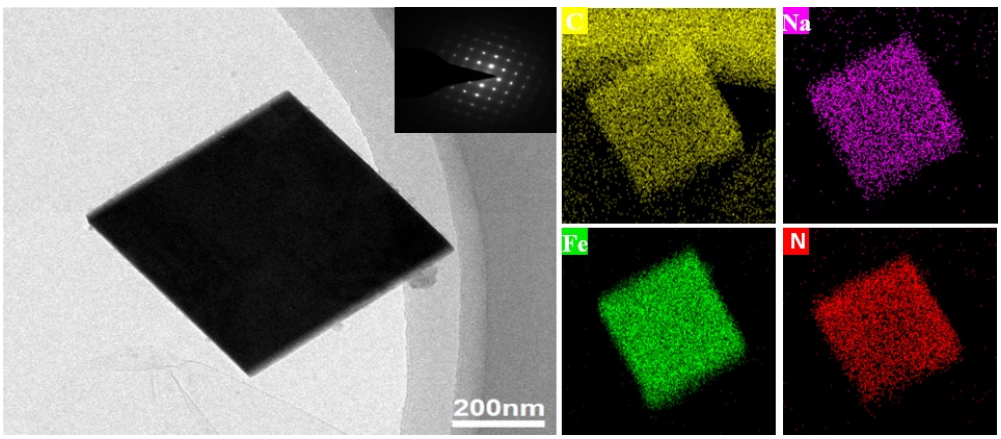

Fig. S2 TEM image of $InsP_6$-PB-10 and the corresponding EDX mapping images.

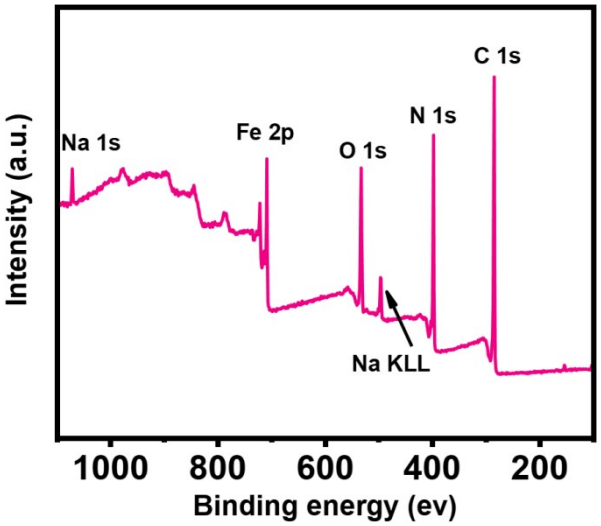

Fig. S3 XPS spectrum of $InsP_6$-PB-10.

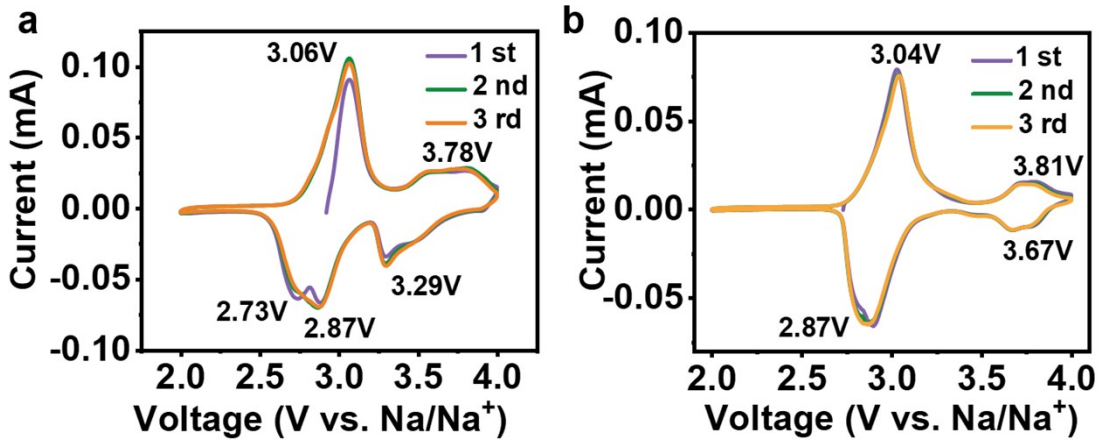

Fig. S4 CV curves at 0.1 mV s^{-1} of (a) $InsP_6$-PB-30, and (b) PB.

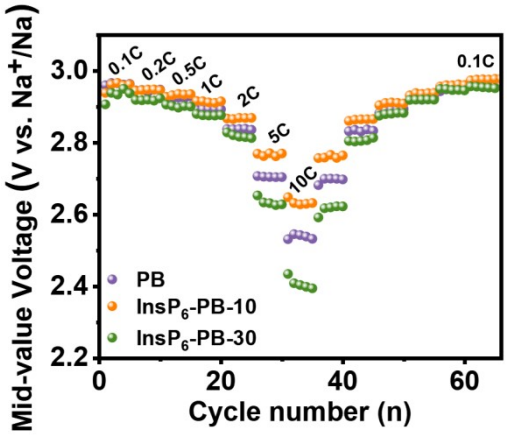

Fig. S5 Mid-value voltage of the samples at different current densities.

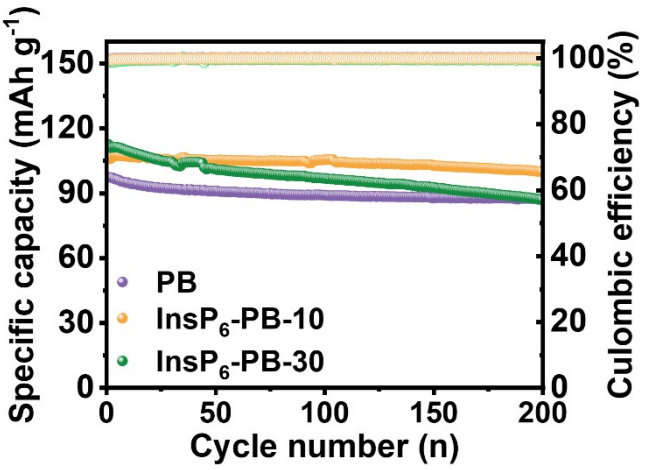

Fig. S6 Cycling performance at 1 C.

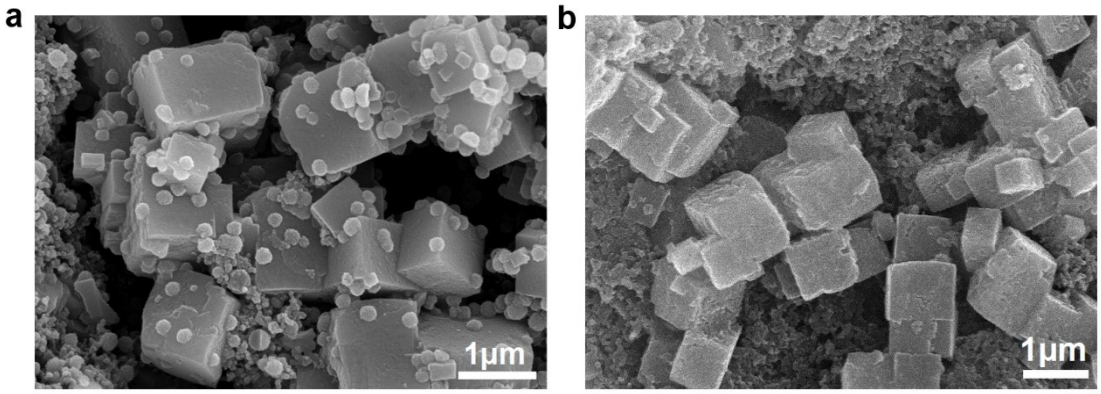

Fig. S7 SEM images of $InsP_6$-PB-10 (a) before, and (b) after 200 cycles.

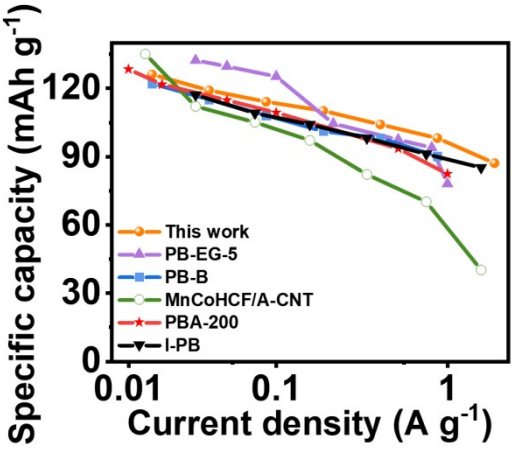

Fig. S8 The rate performance comparison with the recent PB electrodes.

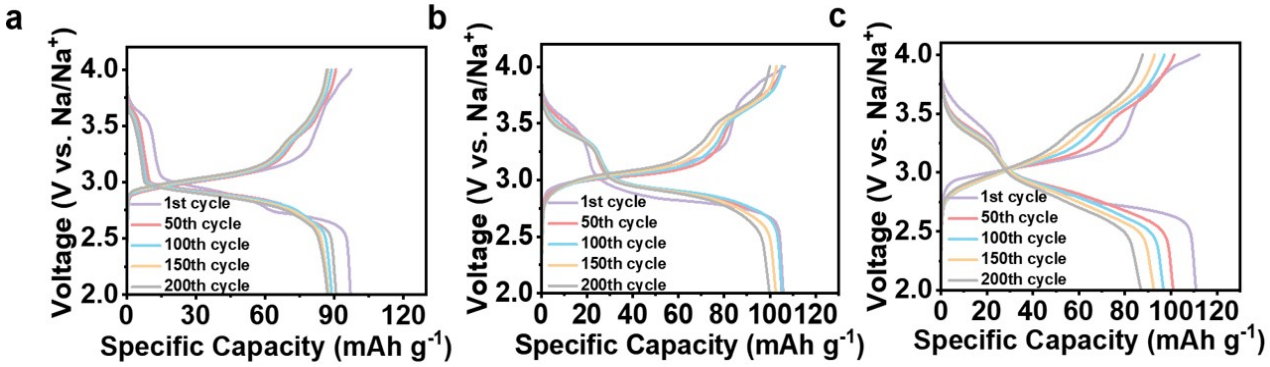

Fig. S9 Charge-discharge profiles at 1C of (a) PB, (b) $InsP_6$-PB-10, and (c) $InsP_6$-PB-30.

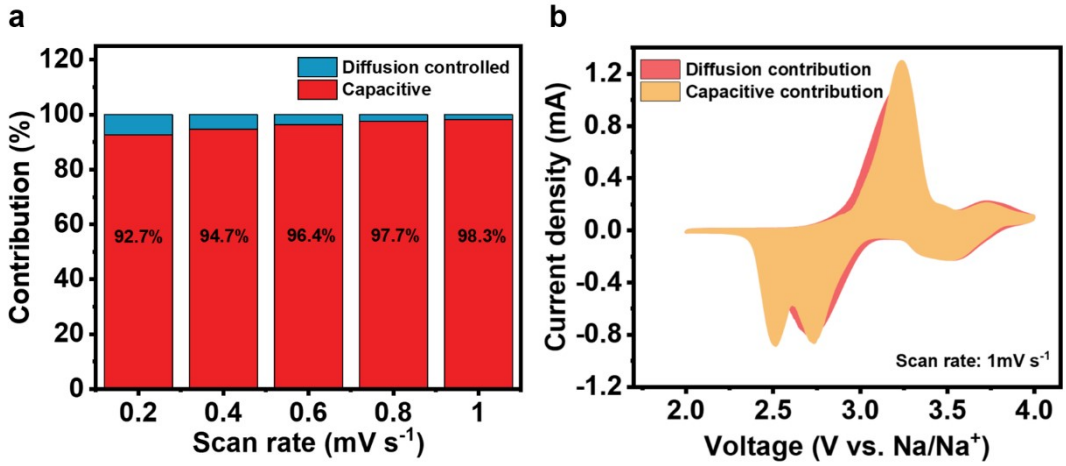

Fig. S10 (a) The proportion of the capacitance contribution at various scanning rates for $InsP_6$-PB-10. (b) Pseudocapacitance determination of $InsP_6$-PB-10 at 1 mV s^{-1}.

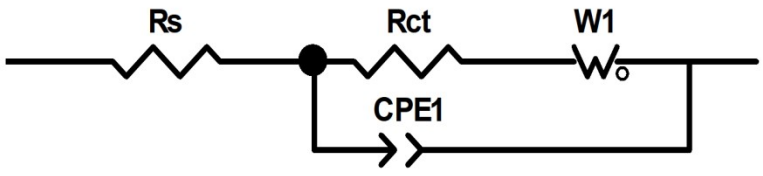

Fig. S11 The circuit model.

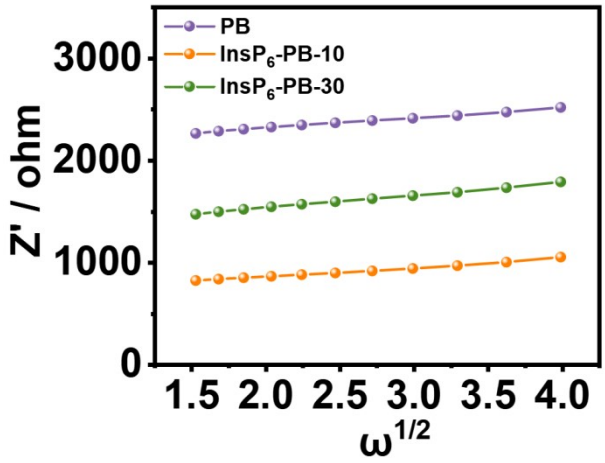

Figure S12. *Z' vs.* $\omega^{-1/2}$ plots.

Table S1 Rietveld-refined structural parameters for the PB sample.

Cubic, space group Fm-3m, a=b=c=10.211 Å, V=1064.63 Å^3.					
Atom	x	y	z	Occupation	Uiso
Na	0.25000	0.25000	0.25000	0.2450	0.02957
Fe1	0.00000	0.00000	0.00000	0.8805	0.01274
Fe2	0.00000	0.00000	0.50000	1.0000	0.01143
C	0.00000	0.00000	0.17788	0.8805	0.01209
N	0.00000	0.00000	0.23859	0.8805	0.10545
O1	0.00000	0.00000	0.23859	0.0260	0.10565
O2	0.31059	0.31059	0.31059	0.0492	0.10085

Table S2 Rietveld-refined structural parameters for $InsP_6$-PB-10.

Cubic, space group Fm-3m, a=b=c=10.205 Å, V=1062.89 Å^3.					
Atom	x	y	z	Occupation	Uiso
Na	0.25000	0.25000	0.25000	0.2850	0.02960
Fe1	0.00000	0.00000	0.00000	0.9694	0.00764
Fe2	0.00000	0.00000	0.50000	1.0000	0.01439
C	0.00000	0.00000	0.17788	0.9694	0.01254
N	0.00000	0.00000	0.23859	0.9694	0.10540
O1	0.00000	0.00000	0.23859	0.0065	0.10560
O2	0.31059	0.31059	0.31059	0.0123	0.10080

Table S3 Rietveld-refined structural parameters for $InsP_6$-PB-30.

Cubic, space group Fm-3m, a=b=c=10.209 Å, V=1064.25 Å^3.					
Atom	x	y	z	Occupation	Uiso
Na	0.25000	0.25000	0.25000	0.2400	0.02960
Fe1	0.00000	0.00000	0.00000	0.9011	0.01270
Fe2	0.00000	0.00000	0.50000	1.0000	0.00715
C	0.00000	0.00000	0.17788	0.9011	0.01210
N	0.00000	0.00000	0.23859	0.9011	0.10540
O1	0.00000	0.00000	0.23859	0.0230	0.10565
O2	0.31059	0.31059	0.31059	0.0470	0.10085

Table S4 Composition analysis of PB, $InsP_6$-PB-10 and $InsP_6$-PB-30.

Sample	**Na**	**Fe**	**C**	**N**
PB	3.71%	34.73%	21.23%	24.55%
$InsP_6$-PB-10	3.91%	33.01%	21.71%	24.46%
$InsP_6$-PB-30	3.38%	32.71%	20.84%	23.34%

Table S5 The electrochemical performance of different types of chelating agents.

Samples	Reversible capacities	Rate capacities	Cycling performance	Reference
Acid–sodium acetate	113 mAh/g at 0.017 A/g	90 mAh/g at 0.85 A/g	81% after 500 cycles at 0.85 A/g	Ref.1[1]
Benzoic acid	110 mAh/g at 0.1 A/g	95.9 mAh/g at 1 A/g	76% after 500 cycles at 1 A/g	Ref.2[2]
Sodium citrate	116 mAh/g at 0.01 A/g	70 mAh/g at 2 A/g	71% after 500 cycles at 0.1 A/g	Ref.3[3]
Sodium polyacrylate	140 mAh/g at 0.034 A/g	105 mAh/g at 5.1 A/g	94.6% after 1000 cycles at 1.7 A/g	Ref.4[4]
Sodium pyrophosphate	128.5 mAh/g at 0.034 A/g	117.6 mAh/g at 0.85 A/g	80.6% after 100 cycles at 0.17 A/g	Ref.5[5]
Cysteine	149.13 mAh/g at 0.025 A/g	60.09 mAh/g at 2 A/g	78.34% after 500 cycles at 0.5 A/g	Ref.6[6]
Na_2EDTA	120 mAh/g at 0.025 A/g	73 mAh/g at 1 A/g	80% after 1300 cycles at 0.5 A/g	Ref.7[7]
Sodium carboxymethyl cellulose	130 mAh/g at 0.05 A/g	116 mAh/g at 1 A/g	97.4% after 3000 cycles at 10 A/g	Ref.8[8]
Inositol hexakisphosphate	127 mAh/g at 0.017 A/g	87 mAh/g at 1.7 A/g	90% after 1000 cycles at 0.85 A/g	This work

Table S6 The electrochemical performance comparison between $InsP_6$-PB-10 and previously reported PB cathode

Samples	Reversible capacities	Rate capacities	Cycling performance	Reference
PB-B	113 mAh/g at 0.017 A/g	90 mAh/g at 0.85 A/g	81% after 500 cycles at 0.85 A/g	Ref.1[1]
BTA-PB-1.6	110 mAh/g at 0.1 A/g	95.9 mAh/g at 1 A/g	76% after 500 cycles at 1 A/g	Ref.2[2]
PB-EG-5	142 mAh/g at 0.01 A/g	91.3 mAh/g at 1 A/g	70% after 1000 cycles at 1 A/g	Ref.9[9]
MnCoHCF/A-CNT	114 mAh/g at 0.03 A/g	94 mAh/g at 0.75 A/g	50.5% after 800 cycles at 0.75 A/g	Ref.10[10]
PBA-200	129 mAh/g at 0.01 A/g	82.3 mAh/g at 1.5 A/g	77% after 500 cycles at 0. 5 A/g	Ref.11[11]
$InsP_6$-PB-10	127 mAh/g at 0.017 A/g	87 mAh/g at 1.7 A/g	90% after 1000 cycles at 0.85 A/g	This work

Table S7 Rs and Rct derived from Nyquist plots of as-prepared samples.

Sample	Rs (Ω)	Rct (Ω)
PB	8.05	1828
$InsP_6$-PB-10	6.21	606
$InsP_6$-PB-30	5.76	1058

Reference

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