**Supporting information**

# Crystal structure engineering of prussian blue through double-acting chelating agent for energy storage devices

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# Equal Contribution;

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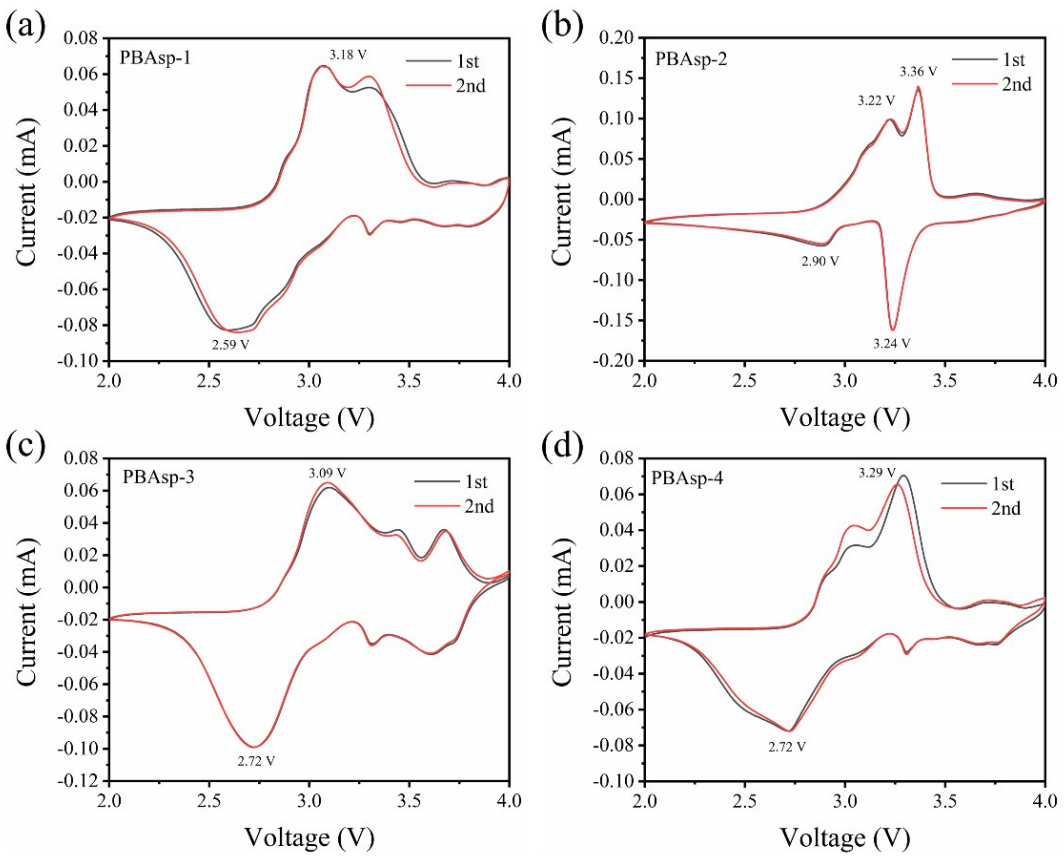


**Fig. S1.** CV curves of the first two cycles for (a) PBAsp-1, (b) PBAsp-2, (c) PBAsp-3 and (d) PBAsp-4 electrodes from 2.0 to 4.0 V at a scan rate of 0.1 mV $s^{-1}$.

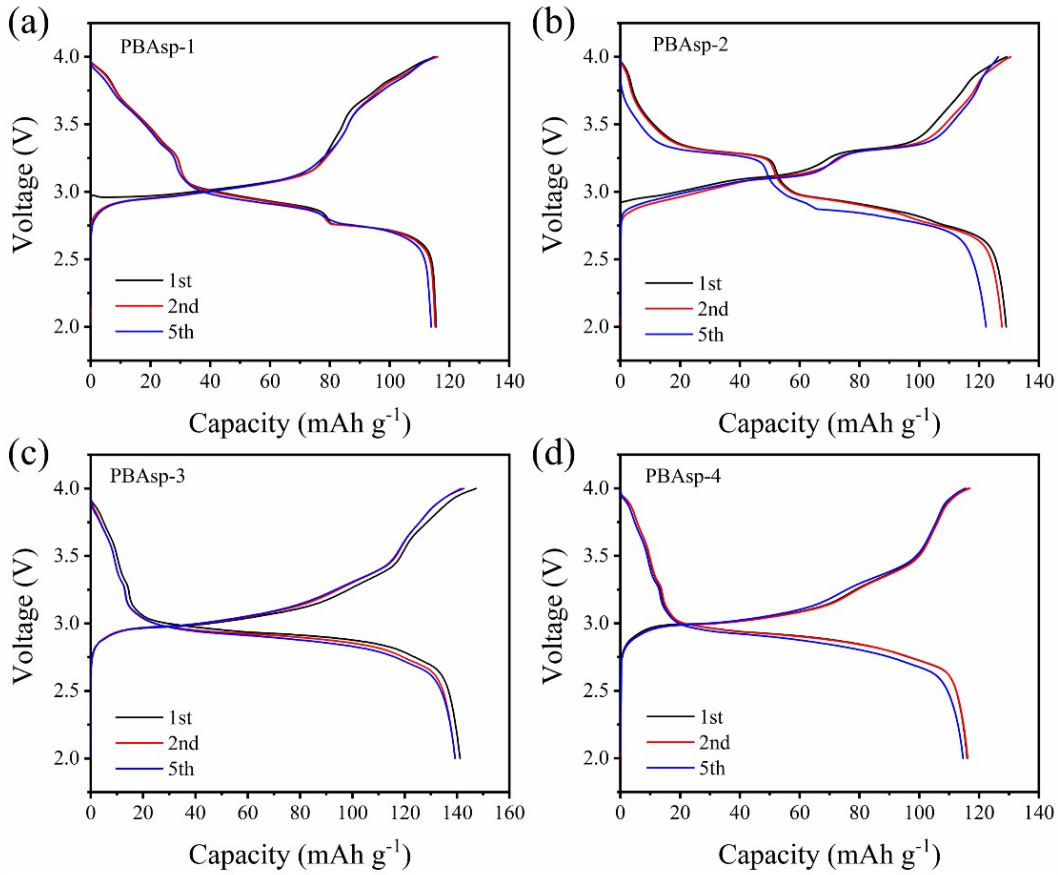


**Fig. S2.** Charge and discharge curves of first three cycles for from 2.0 to 4.0 V at 50mA $g^{-1}$: (a) PBAsp-1, (b) PBAsp-2, (c) PBAsp-3, (d) PBAsp-4.

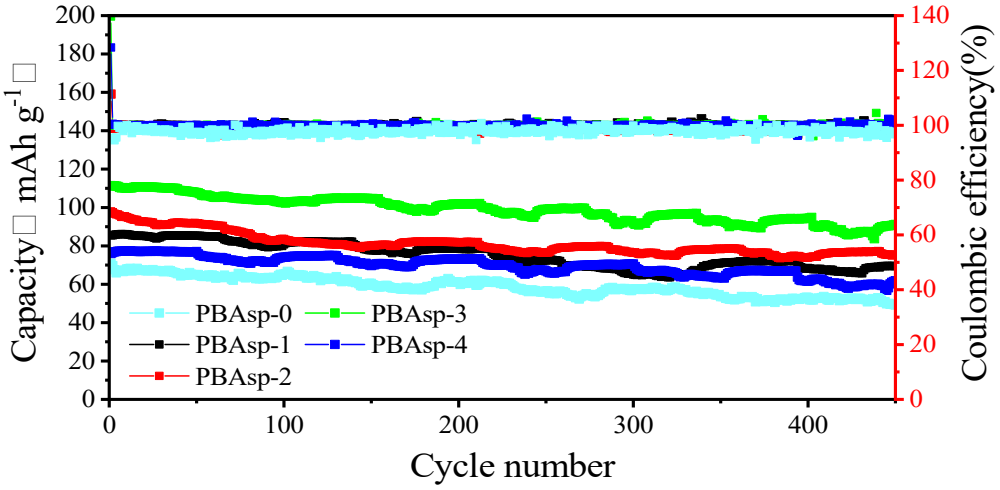


**Fig. S3.** Cycle performance of PBAsp-0; PBAsp-1; PBAsp-2; PBAsp-3 and PBAsp-4 electrodes at 500 mA $g^{-1}$ in the voltage range of 2.0-4.0 V.

**Table S1.** Ratio of $Fe^{2+}$ and $Fe^{3+}$ in all samples

| Samples | $Fe^{2+}$ | $Fe^{3+}$ |
|---|---|---|
| PBAsp-1 | 31.4% | 69.6% |
| PBAsp-2 | 48.5% | 51.5% |
| PBAsp-3 | 58.2% | 41.8% |
| PBAsp-4 | 57.0% | 43.0% |

**Table S2.** The calculated resistance based on the Nyquist curves of as-prepared samples.

| Samples | $R_S$ (Ω) | $R$ct (Ω) |
|---|---|---|
| PBAsp-1 | 18.18 | 1539 |
| PBAsp-2 | 13.77 | 1076 |
| PBAsp-3 | 11.09 | 963.1 |
| PBAsp-4 | 4.82 | 215.6 |

**Table S3.** Comparison of the performance of various chelating agents for the preparation of Prussian blue.

| Samples | Utilization rate | Capacity(1C) | Cycle life |
|---|---|---|---|
| this work | 4.4 g | 131 mAh $g^{-1}$ | 81.81%, 450 (5C) |
| sodium citrate[1] | 13.04 g | 100 mAh $g^{-1}$ | 71%, 500 (1C) |
| sodium pyrophosphate[2] | 26.36 g | 124 mAh $g^{-1}$ | 80.6%, 100 (1C) |
| EDTA[3] | 11.2 g | 151 mAh $g^{-1}$ | 84%, 100 (1C) |

**References**


1 S. Lim, D. Choi, T. Jeong and D. Han, *J. Alloys Compd.* , 2023, **938**, 168502.

2 J. Peng, J. Huang, Y. Gao, Y. Qiao, H. Dong, Y. Liu, L. Li, J. Wang, S. Dou and S. Chou, *Small*, 2023, **19**, 2300435.

3 Z.G. Neale, C. Liu and G. Cao, *Sustain. Energy Fuels*, 2020, **4**, 2884−2891.

**Table S4.** Comparison of the cost of using aspartic acid and sodium citrate.

| Samples | Price (RMB/ton) | Utilization rate (g) | Usage cost (RMB) |
|---|---|---|---|
| aspartic acid | 25000 | 4.4 | 0.11 |
| sodium citrate | 5100 | 13.04 | 0.067 |

Note: The above calculations are based on equal masses of sodium ferrocyanide, prepared to produce the same yield of Prussian blue obtained.