

Supplementary Information

Regulating Electric Double Layer Dynamics for Robust SEI in Fast-Charging Graphite Anodes

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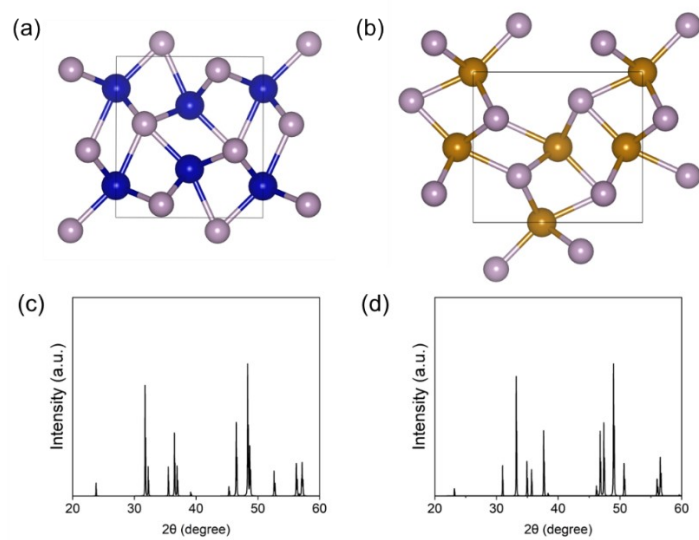


Fig. S1 Calculated crystal structures and XRD peaks of (a, c) CoP and (b, d) FeP, respectively.

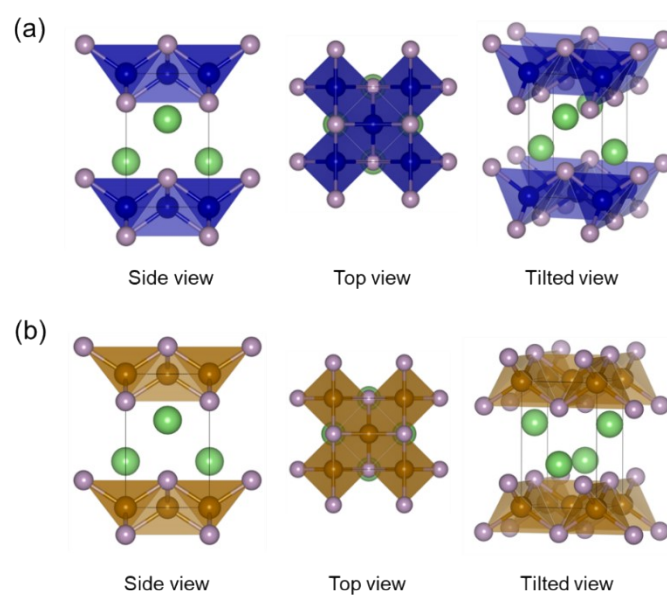


Fig. S2 Calculated crystal structures of (a) LiCoP and (b) LiFeP different viewing angles.

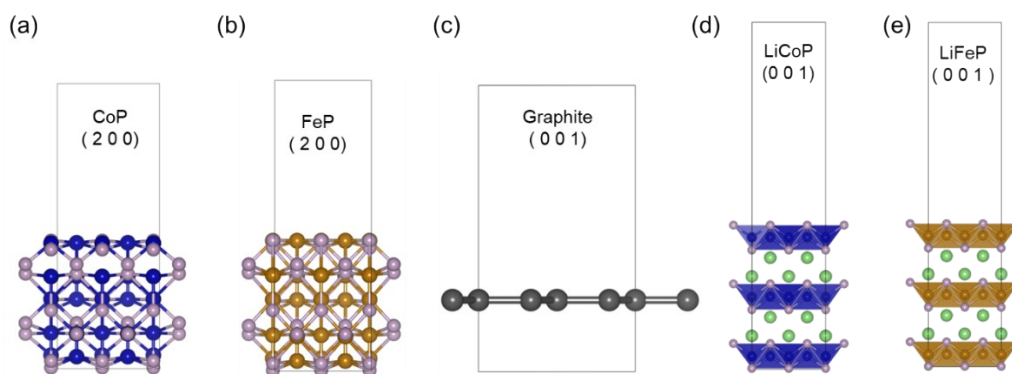


Fig. S3 The calculated surface of (a) (200) for CoP, (b) (200) for FeP, (c) (001) for graphite, (d) (001) for LiCoP, and (e) (001) LiFeP.

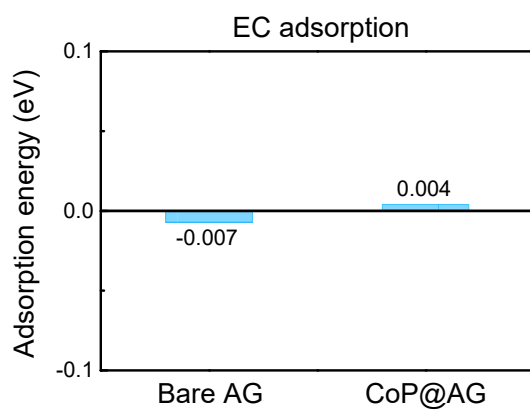


Fig. S4 Adsorption energy of EC for Bare AG and CoP@AG surface.

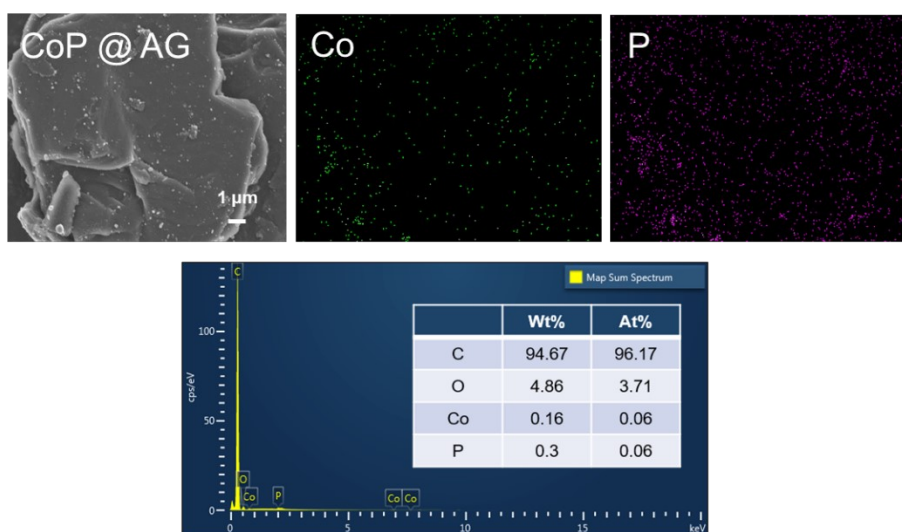


Fig. S5 Elemental distribution and composition ratio of CoP@AG

| Sample | Element | Result | |
|--------|---------|---------|---------------------|
| CoP@AG | Co | 0.45wt% | ICP-OES iCAP Pro |
| | P | 0.3wt% | |

Fig. S6 ICP-OES measurement of CoP@AG powder

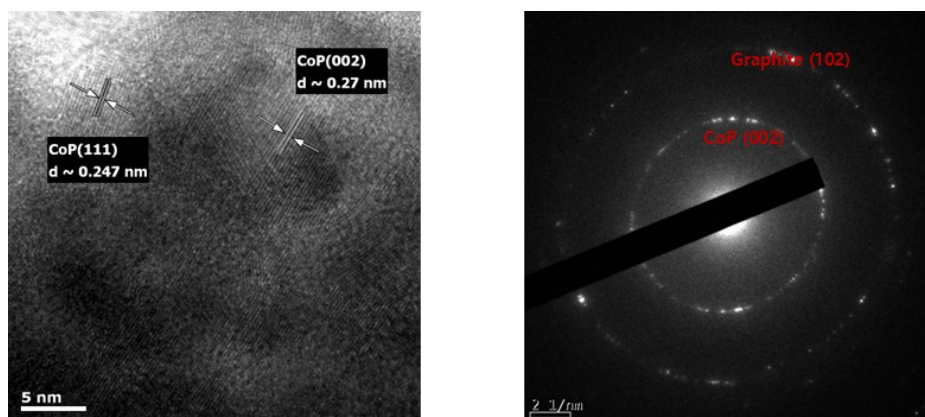


Fig. S7 TEM and SAED images of CoP@AG particles

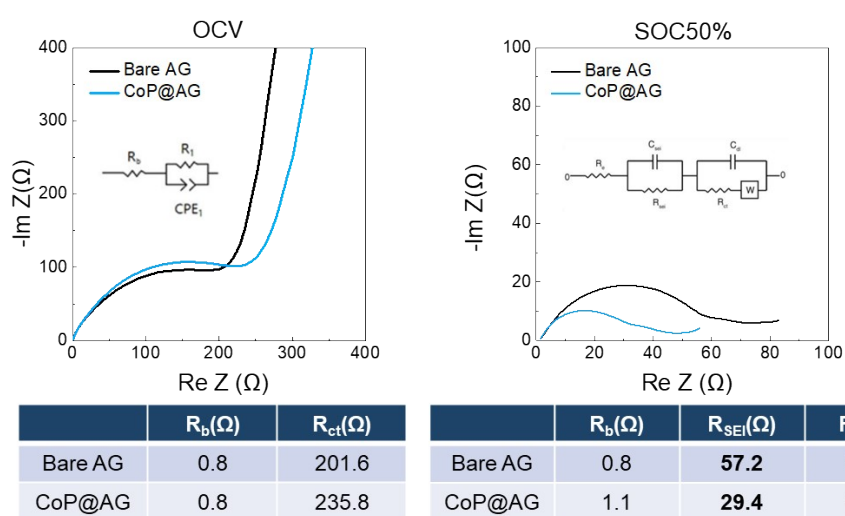


Fig. S8 Nyquist plots of Bare AG and CoP@AG half cells at OCV state and SOC 50% state.

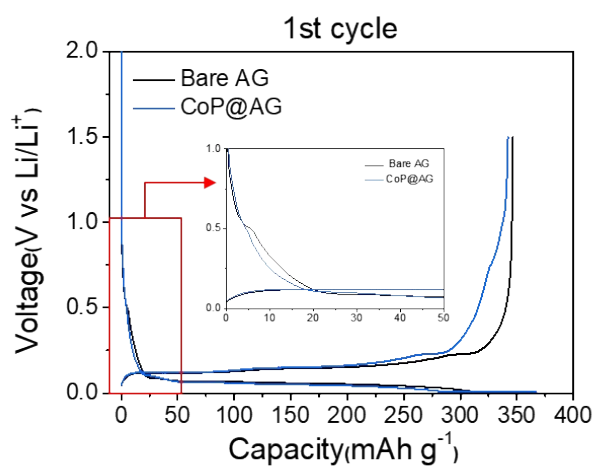


Fig. S9 Galvanostatic charge/discharge profiles of Bare AG and CoP@AG using electrolyte without FEC additive.

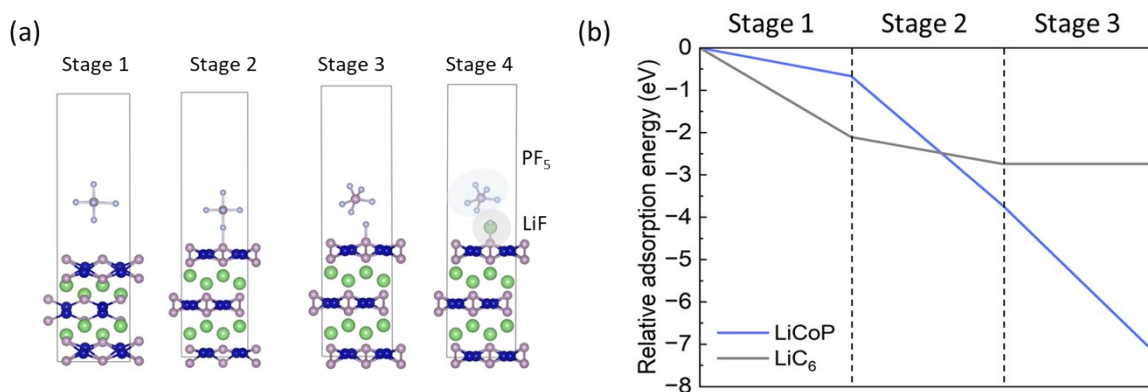


Fig. S10 (a) Decomposition process of PF_6^- and formation of LiF on LiCoP surfaces. (b) The relative adsorption (formation) energies of PF_6^- adsorption (Stage 1), PF_6^- decomposition (Stage 2), PF_6^- decomposition (Stage 3) and LiF formation (Stage 4) on LiCoP and LiC_6 surface.

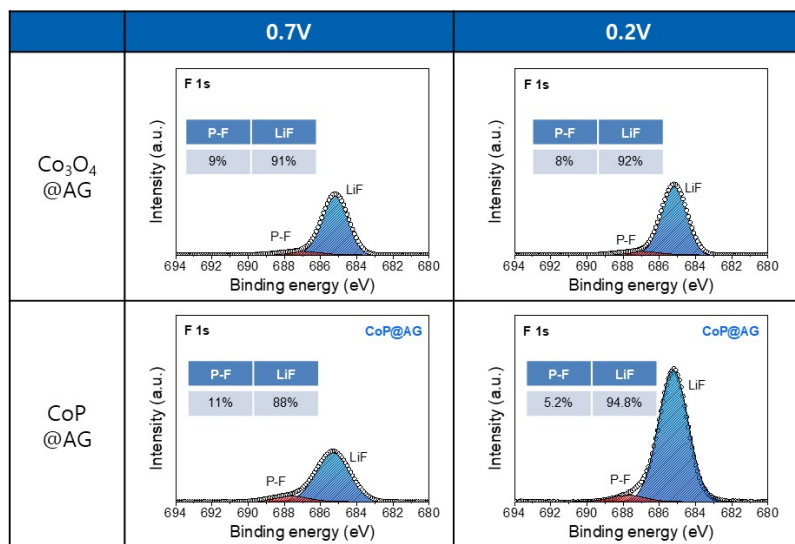


Fig. S11 Evolution behavior of the SEI layer of Co₃O₄@AG through XPS analysis after charging to 0.7V and 0.2V.

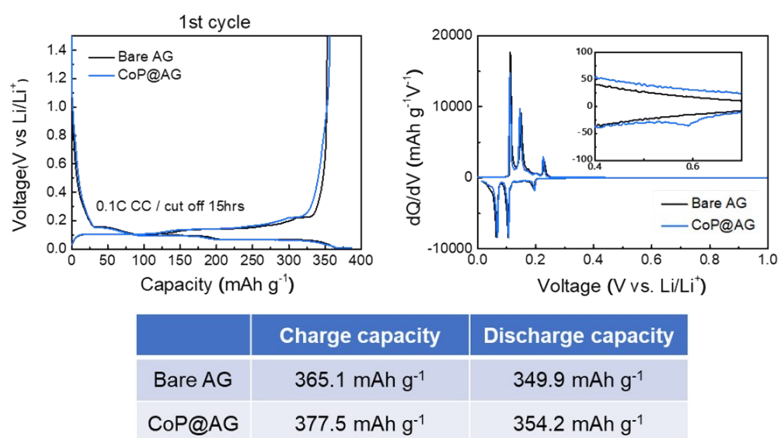


Fig. S12 Galvanostatic charge/discharge profiles and dQ/dV plot of Bare AG and CoP@AG using electrolyte that containing FEC additive.

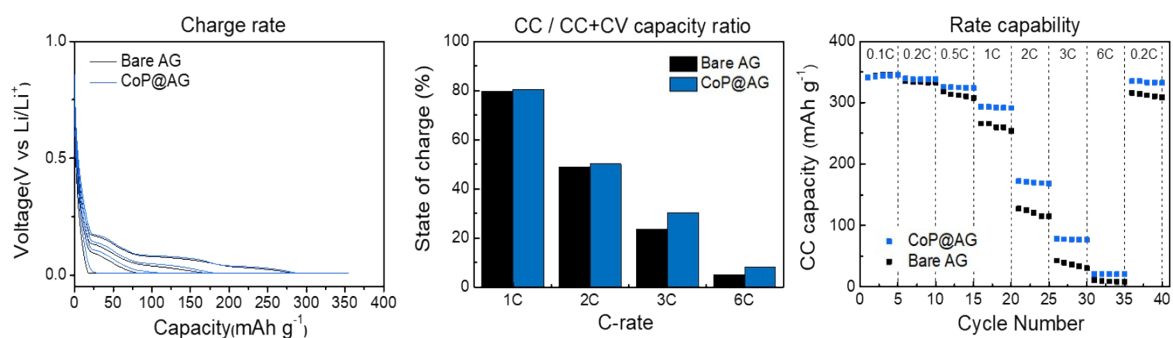


Fig. S13 Rate capability of the Bare AG and CoP@AG half-cell at various C-rates.

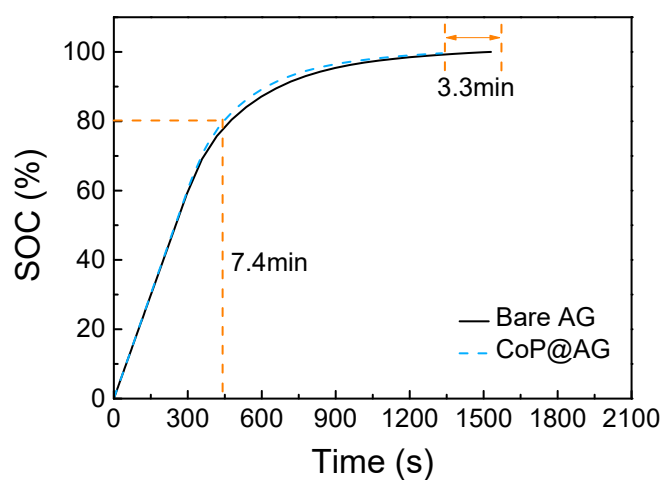
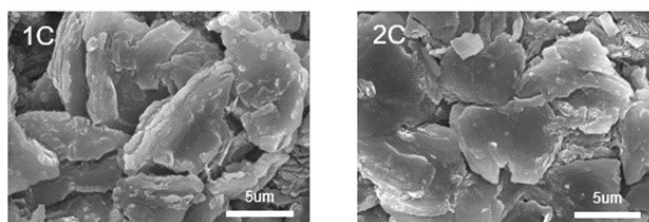


Fig. S14 SOC vs. time profiles of full cells with Bare AG and CoP@AG anodes.

NCM811 / Bare AG



NCM811 / CoP@AG

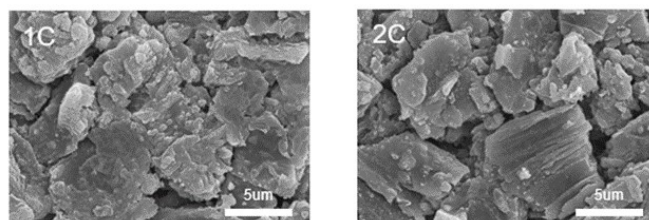


Fig. S15 SEM images of Bare AG and CoP@AG electrodes after 1C, 2C charge.

NCM 811 / Bare AG

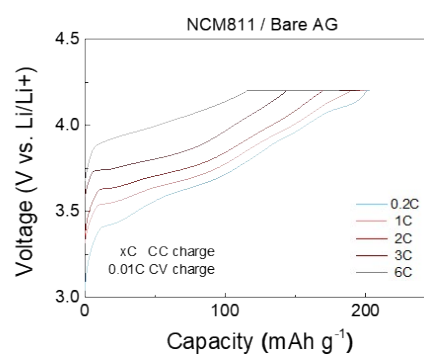
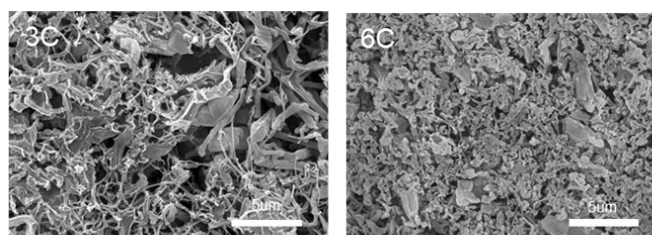
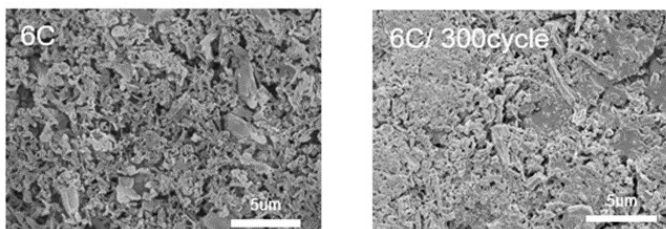


Fig. S16 SEM images of Bare AG electrodes after 3C,6C charge and voltage profile of NCM811 / Bare AG full cell at various C rates.

NCM 811 / Bare AG



NCM 811 / CoP@AG

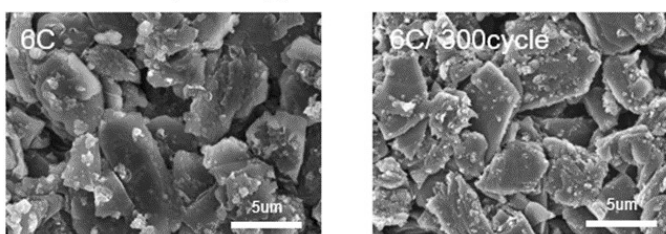
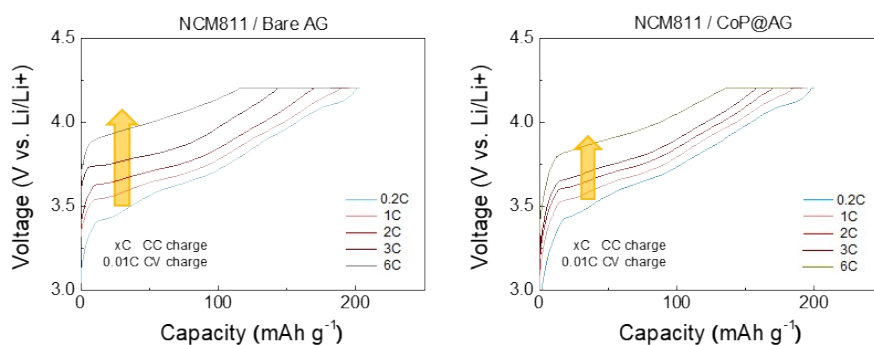


Fig. S17 SEM images of Bare AG and CoP@AG electrodes after 300 cycles at 6C.



| CC/CC+CV(%) | 0.2C | 1C | 2C | 3C | 6C |
|-------------|-------|-------|-------|-------|------|
| Bare AG | 98.7% | 91.9% | 84.1% | 71.4% | 58.7 |
| CoP@AG | 98.7% | 93.0% | 86.4% | 80.6% | 68.0 |

Fig. S18 The CC/CC+CV ratio at each C rate for Bare AG and CoP@AG full cells.

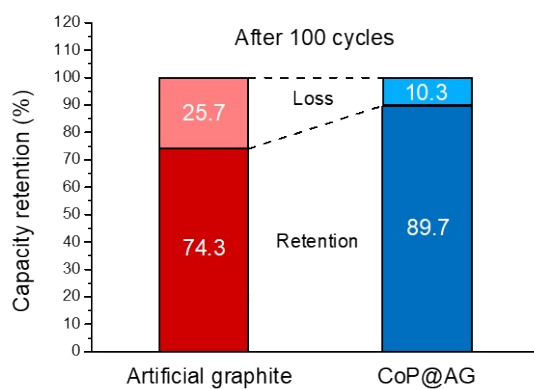


Fig. S19 Capacity retentions of Bare AG and CoP@AG full cells after 100 cycles(6C charging-1C discharging).

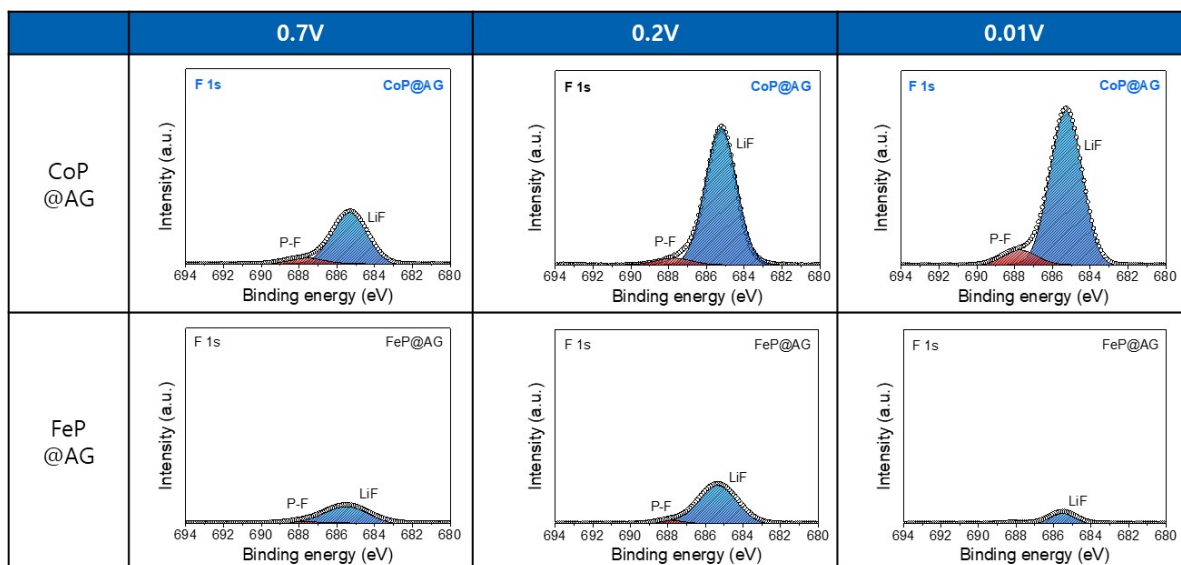


Fig. S20 Evolution behavior of the SEI layer of FeP@AG through XPS analysis after charging to 0.7V, 0.2V and 0.01V.

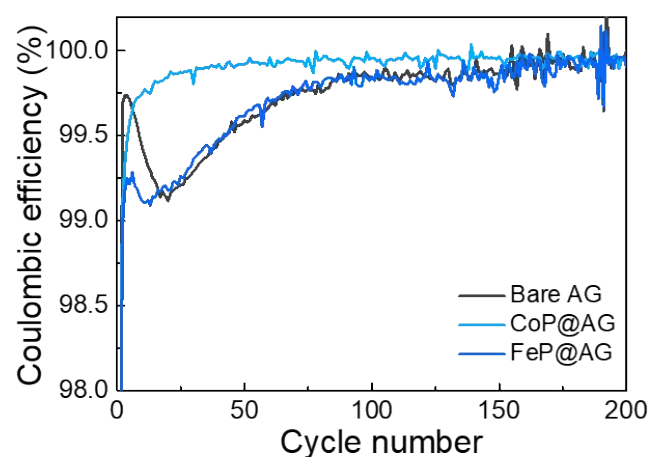


Fig. S21 Coulombic efficiency of Bare AG, CoP@AG and FeP@AG

| Example | Electrode composition(wt%) (AM ^a : CM ^b : BM ^c) | N/P ratio | Areal capacity (mAh cm ⁻²) | Electrode density (g cm ⁻³) | ICE | Fast-charge cyclability (retention) (%cycle no.) | Reference |
|--|---|------------|--|---|-----------|--|------------------|
| GPC | 92:2:6 | 1.1 | 1.5 | - | 91.1 | 1C (84%, 200cycle) | 1 |
| Gr@La_{0.1}TNO | 93:2:5 | 0.6 | 0.95 | - | 86.4 | 3C(76%, 200cycle) | 2 |
| Photo-graphite | 93:3:4 | 1.1 | 2.9 | - | 94.3 | 3C(95%, 50cycle) | 3 |
| Graphite | 80:10:10 | 1 | 0.3 | - | - | 4C(86%,100 cycle) | 4 |
| G@Cu-CuNW | 94:4:2 | 1.05 | 3.2 | 1.5 | 91 | 3C(50%,50c ycle) | 5 |
| Edge-plane activated graphite(EAG) | 96:1:3 | 1.1 | 3.34 | 1.6 | 93.8 | 3C(70%,50c ycle) | 6 |
| Mo-CP/graphite | 96:0:4 | 1.1 | 2.2 | 1.5 | 92 | 6C(84%,300 cycle) | 7 |
| CoP@AG (This work) | 97.5:1:1.5 | 1.1 | 2.2 | 1.5 | 91 | 6C(88%,300 cycle) | This work |

a. Active material, b. Conductive agent, c. Binder material

Supplementary Table 1. Comparison of the fast chargeability of various anode materials for

LIBs

Supplementary References

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2. Y. L. Sheng, X. Y. Yue, W. Hao, Y. T. Dong, Y. K. Liu and Z. Liang, *Nano Lett*, 2024, **24**, 3694-3701.
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