**Supplemental Information**

Upcycling the spent graphite/LiCoO2 batteries for high-voltage graphite/LiCoPO4-co-workable dual-ion batteries

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Fig. S1 (a, b) SEM images of spent LiCoO2 material under different magnifications.

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Fig. S2 (a, b) SEM images of spent graphite material under different magnifications.

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Fig. S3 (a, b) SEM images of spent LiCoO2 material after ball‐milling under different magnifications.

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Fig. S4. CV profile of LCP at 0.1 mV⋅s−1.

图表, 折线图

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Fig. S5. CV profile of G at 0.1 mV⋅s−1.

图示

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Fig. S6. GCD curves of RLCPG and LCP from the current densities of 25 to 1000 mA⋅g−1.

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Fig. S7 Rate capability of G at various current densities from 25 to 1000 mA⋅g−1.

图示

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Fig. S8. (a, c) One single GITT profile of LCP and RLCPG in the discharge processes. (b, d) The corresponding linearly fitting behavior for the *E* vs. *τ* 1/2 in the discharge GITT titration.

The apparent Li+ diffusion coefficients (*D*app,Li) were further calculated by GITT tests. The GITT tests were performed at different states at current density of 5 mA⋅g-1 during the charge-discharge process. The *D*app,Li can be calculated by the following formula [1]:

() (S1)

where *m*, *M*, *L*, and *V* are the mass, molecular weight, average radius, and molar volume of electrode, respectively; *s* is the active surface area of electrode; *τ* is the time for an applied galvanostatic current; *ΔE*s together with *ΔEτ* are the quasi-equilibrium potential and the total change of cell voltage *E* during the current pulse.

**References**

[1] Z.Y. Gu, J.Z. Guo, J.M. Cao, *et al.*, An advanced high-entropy fluorophosphate cathode for sodium-ion batteries with increased working voltage and energy density, *Adv*. *Mater*., 34(2022), No. 14, art. No. 2110108.