Supporting Information for

# Kinetic Limits of Graphite Anode for Fast-Charging Lithium-Ion Batteries

Suting Weng[1, 2], Gaojing Yang[1, 2], Simeng Zhang[1, 3], Xiaozhi Liu[1], Xiao Zhang[1, 3], Zepeng Liu[1, 2], Mengyan Cao[1, 3], Mehmet Nurullah Ateş[4], Yejing Li[1, *], Liquan Chen[1], Zhaoxiang Wang[1, 2, 3, *] and Xuefeng Wang[1, 2, 3, 5, *]

[1] Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing, 100190, P. R. China

[2] School of Physical Sciences, University of Chinese Academy of Sciences, Beijing, 100049, P. R. China

[3] College of Materials Science and Opto-Electronic Technology, University of Chinese Academy of Sciences, Beijing, 100049, P. R. China

[4] Department of Chemistry, Boğazici University, Bebek, Istanbul, 34342, Türkiye

[5] Tianmu Lake Institute of Advanced Energy Storage Technologies Co. Ltd., Liyang, 213300, P. R. China

*Corresponding authors. E-mail: wxf@iphy.ac.cn (X. Wang); zxwang@iphy.ac.cn (Z. Wang); liyejing26@gmail.com (Y. Li)

## Supplementary Figures and Tables

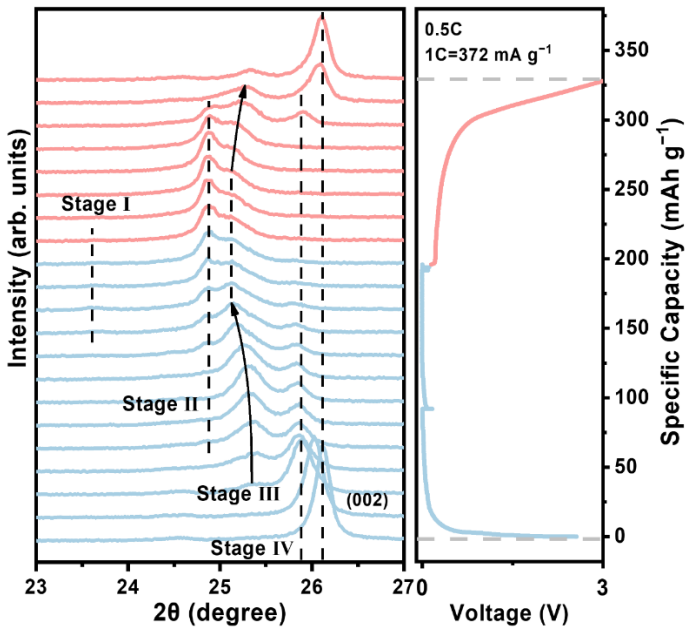


**Fig. S1** The structural evolution of the graphite during electrochemical lithiation and delithiation at 0.5C

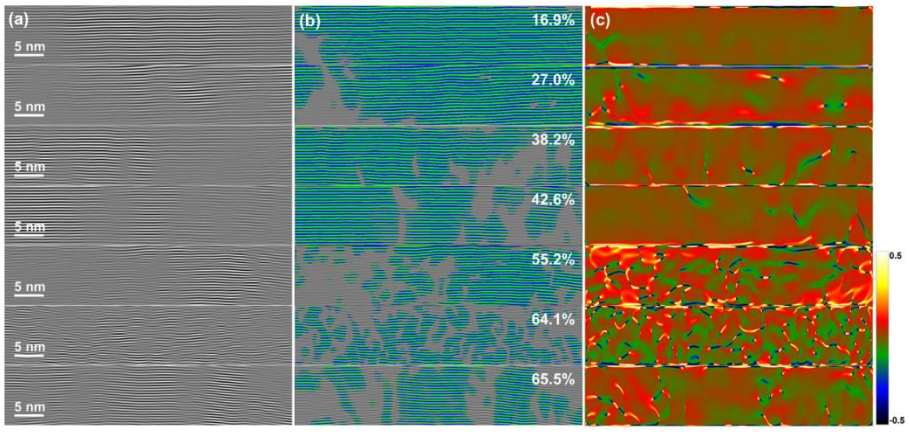


**Fig. S2 a** Inverse fast Fourier transform images, the corresponding **b** area of the defective domains and **c** strain mapping of different graphite regions lithiated at 0.5C. The color from red-to-yellow-to-white in the strain mapping represents the gradually increased tensive strain while that from green-to-blue-to-black indicates the gradually enhanced compressive strain

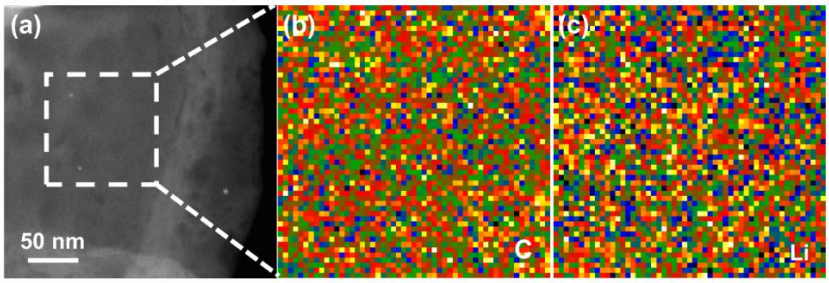


**Fig. S3 a** STEM-ADF image. **b-c** The corresponding EELS mapping of C and Li K-edge of graphite lithiated at 0.05C

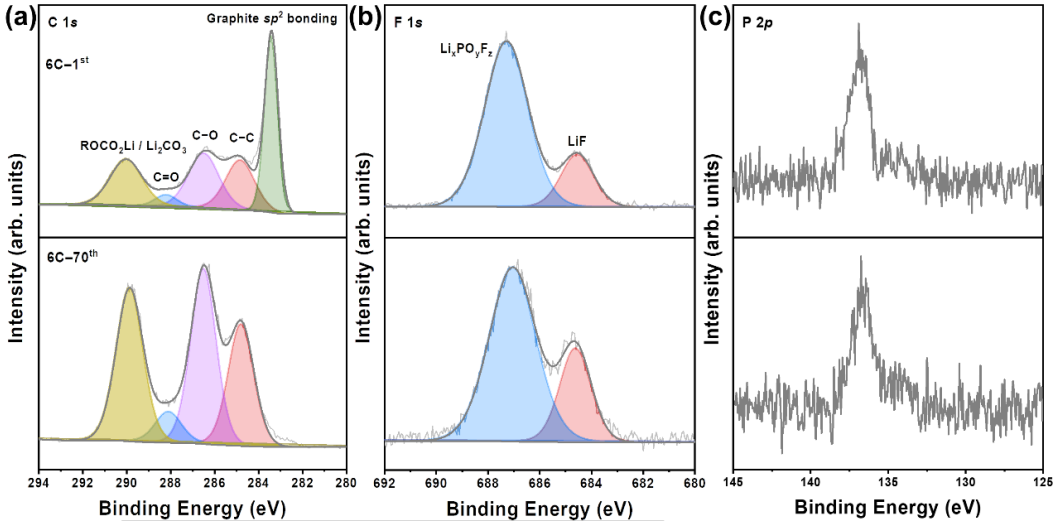


**Fig. S4** XPS spectra of the SEI layers formed on graphite after 1st and 70th cycle at 6C. **a** C 1*s*, **b** F 1*s*, and **c** P 2*p*. The results show that the SEI components change slightly (Table S1) while it becomes thicker since the $sp^2$ bonding signal around 283.5 eV from

the graphite bulk is absent after 70 cycles (Fig. S4a)

**Table S1** The relative content of specific species in the SEI layer based on the XPS spectra in Fig. S4

| Species | 6C-1$^{st}$ | 6C-70$^{th}$ |
|---|---|---|
| C−C | 29.50% | 24.77% |
| C−O | 34.49% | 35.68% |
| C=O | 5.14% | 6.36% |
| $ROCO_2Li/Li_2CO_3$ | 30.87% | 33.19% |
| LiF | 19.04% | 26.24% |
| $Li_xPO_yF_z$ | 80.96% | 73.76% |

**Table S2** Lithium content and its corresponding chemical diffusion coefficient of graphite at different discharge potentials

| Potential (V *vs.* $Li^+/Li^0$) | 0.198 | 0.160 | 0.110 | 0.100 | 0.075 | 0.073 | 0 |
|---|---|---|---|---|---|---|---|
| x in $Li_xC_6$ | 0.025 | 0.127 | 0.231 | 0.376 | 0.531 | 0.703 | 1 |
| D ($10^{-10}$ $cm^2$ $s^{-1}$) | 19.87 | 4.02 | 2.81 | 5.01 | 2.64 | 3.45 | 3.25 |

**Table S3** Lithium content and its corresponding chemical diffusion coefficient of graphite at different charge potentials

| Potential (V *vs.* $Li^+/Li^0$) | 0.700 | 0.245 | 0.200 | 0.165 | 0.135 |
|---|---|---|---|---|---|
| x in $Li_xC_6$ | 0.096 | 0.139 | 0.217 | 0.288 | 0.551 |
| D ($10^{-10}$ $cm^2$ $s^{-1}$) | 5.59 | 6.61 | 11.80 | 10.30 | 11.39 |

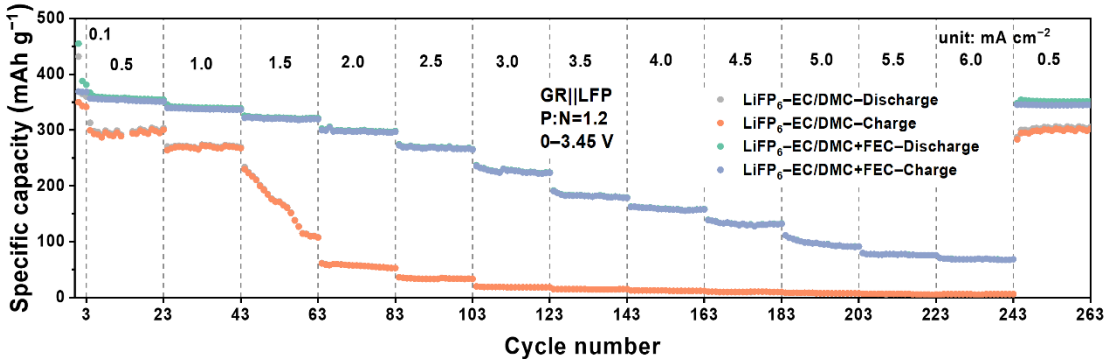


**Fig. S5** The rate performances of graphite||$LiFePO_4$ cells

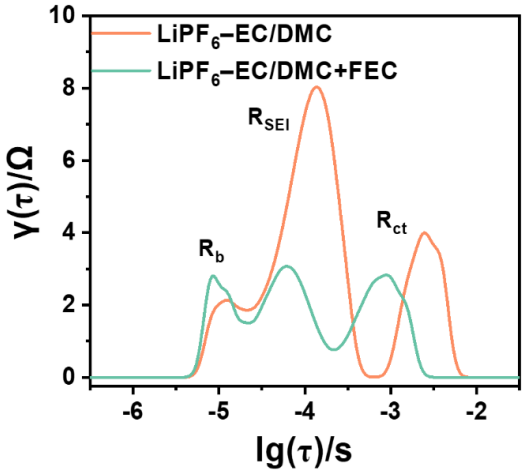


**Fig. S6** The corresponding distribution of relaxation times (DRT) analysis of the electrochemical impedance spectra in Fig. 5b

**Table S4** The corresponding $R_b$, $R_{SEI}$, and $R_{ct}$ values obtained by integrating the natural logarithm ln(τ) of the time constant τ with γ(τ)

| Resistance (Ω) | $LiPF_6$–EC/DMC | $LiPF_6$–EC/DMC+FEC |
|---|---|---|
| $R_b$ | 2.35 | 2.79 |
| $R_{SEI}$ | 13.77 | 4.67 |
| $R_{ct}$ | 4.77 | 4.44 |

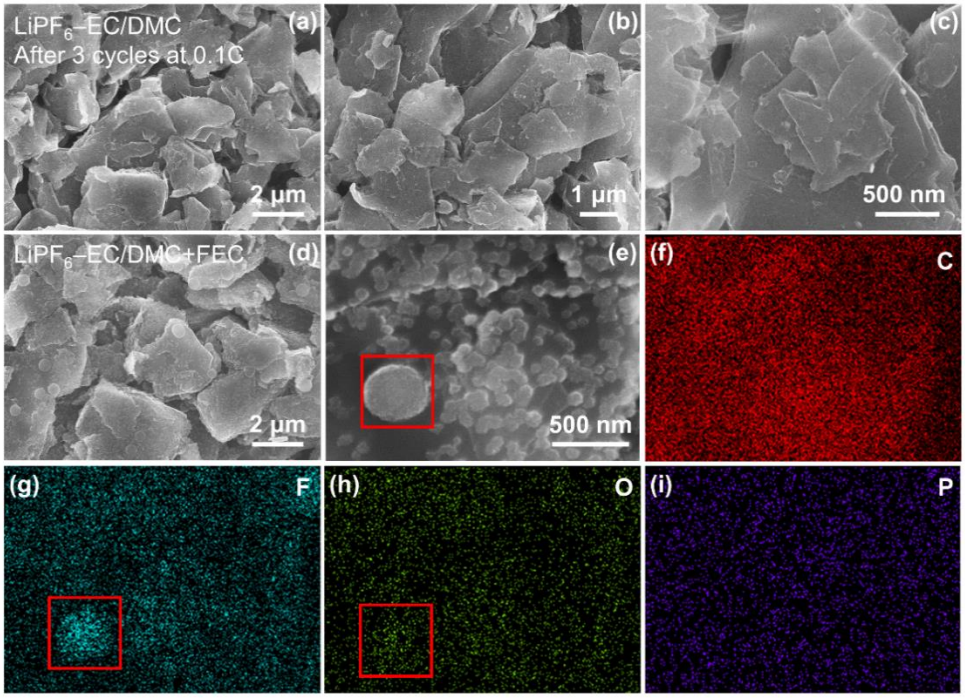


**Fig. S7** SEM images with different magnifications of graphite cycled in **a-c** $LiFP_6$–EC/DMC and **d-e** $LiFP_6$–EC/DMC+FEC electrolytes after 3 cycles at 0.1C. **f-i** The corresponding elements distribution of **e**

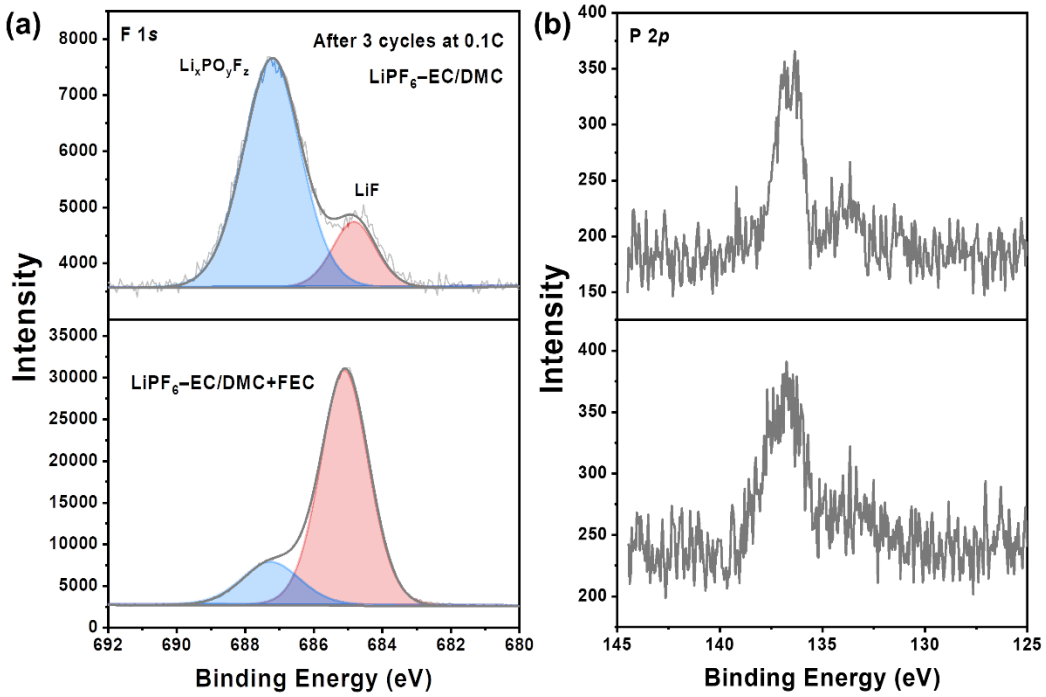


**Fig. S8** XPS spectra of the SEI layers formed on graphite in $LiFP_6$–EC/DMC and $LiFP_6$–EC/DMC+FEC electrolytes after 3 cycles at 0.1C. **a** F 1*s* and **b** P 2*p*

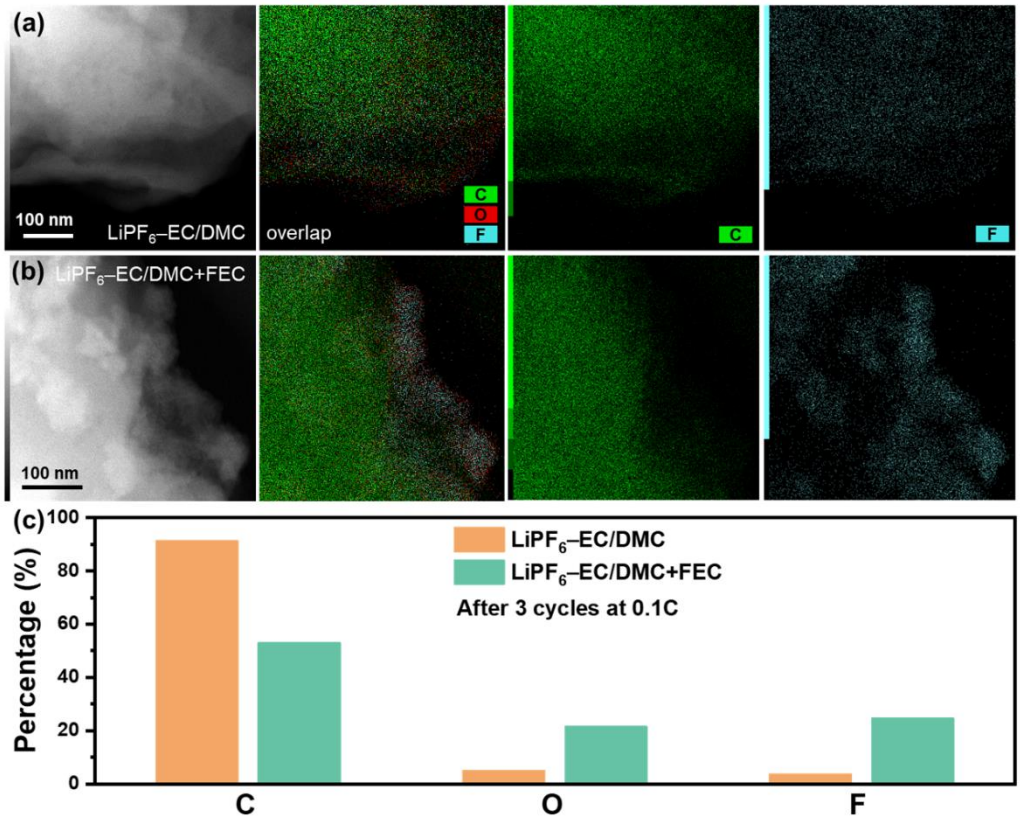


**Fig. S9** EDS mapping of SEI layers formed on graphite in **a** $LiFP_6$–EC/DMC and **b** $LiFP_6$–EC/DMC+FEC electrolytes after 3 cycles at 0.1C. **c** Corresponding different elemental percentage of SEI layers

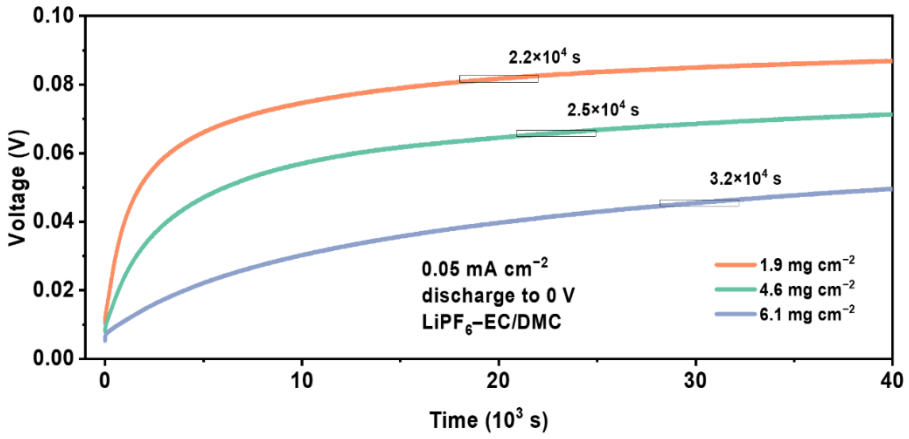


**Fig. S10** Potential relaxation profiles of graphite electrodes with different thicknesses discharged to 0 V at the same areal current density

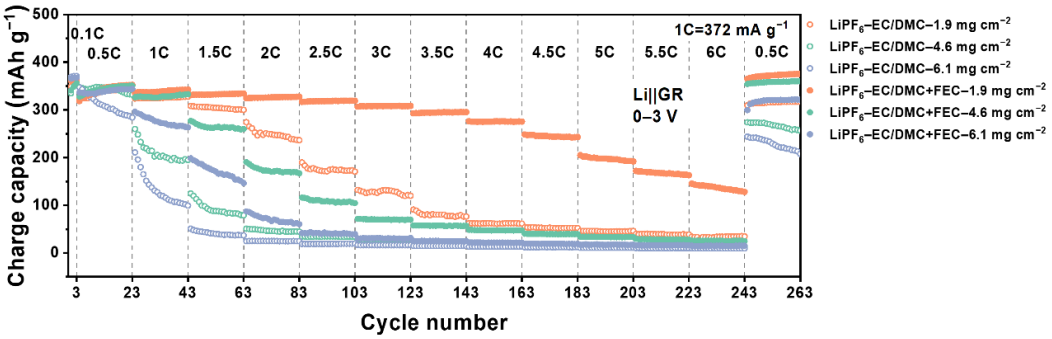


**Fig. S11** The rate performances of different thickness graphite electrodes in Li||graphite cells using $LiFP_6$–EC/DMC and $LiFP_6$–EC/DMC+FEC electrolytes

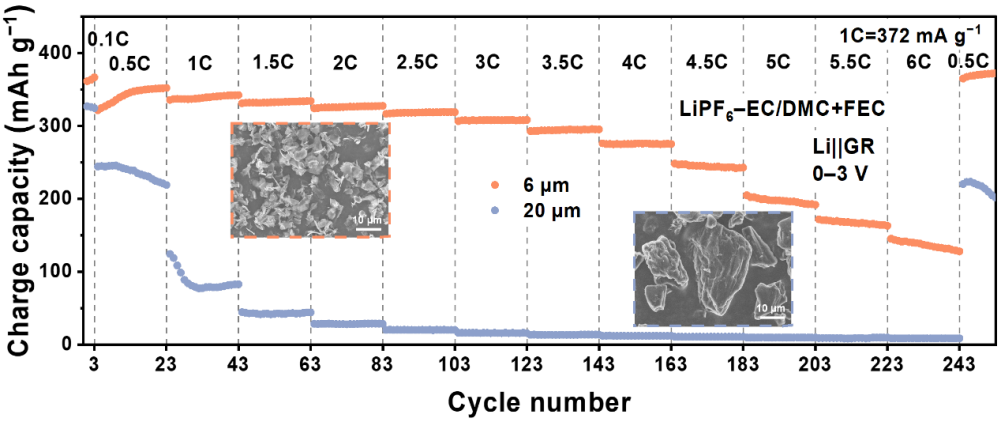


**Fig. S12** The rate performances of Li||graphite cells with $LiFP_6$–EC/DMC+FEC electrolytes using different particle sizes of graphite