

## Supporting Information

### Resolving PC-Graphite Incompatibility via Steric-Hindrance-Directed Interfacial Regulation

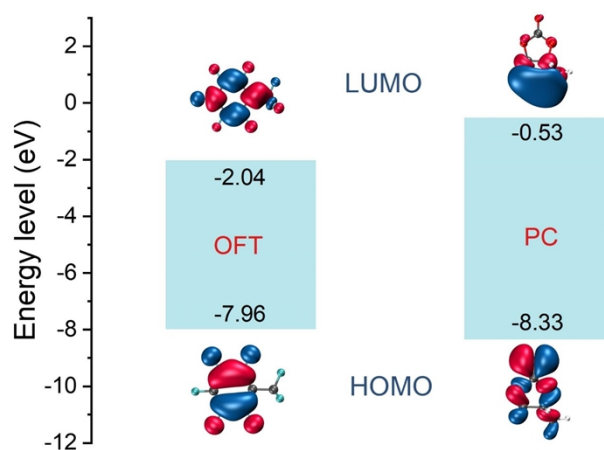
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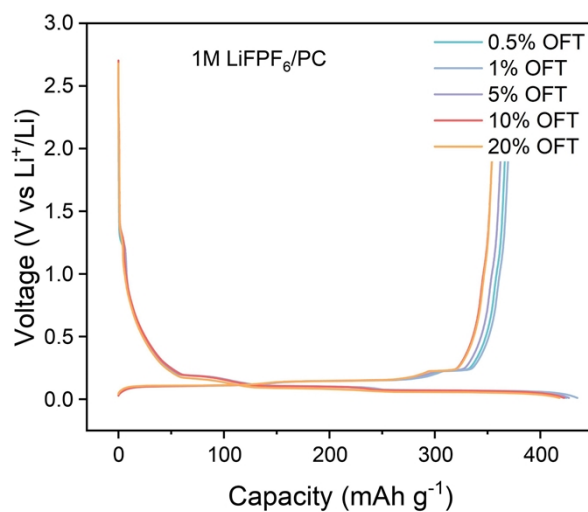
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Molecular Science, Wuhan University, Wuhan 430072, Hubei, China

<sup>#</sup> Huayi Qian and Yongkang Wei contributed equally to this work

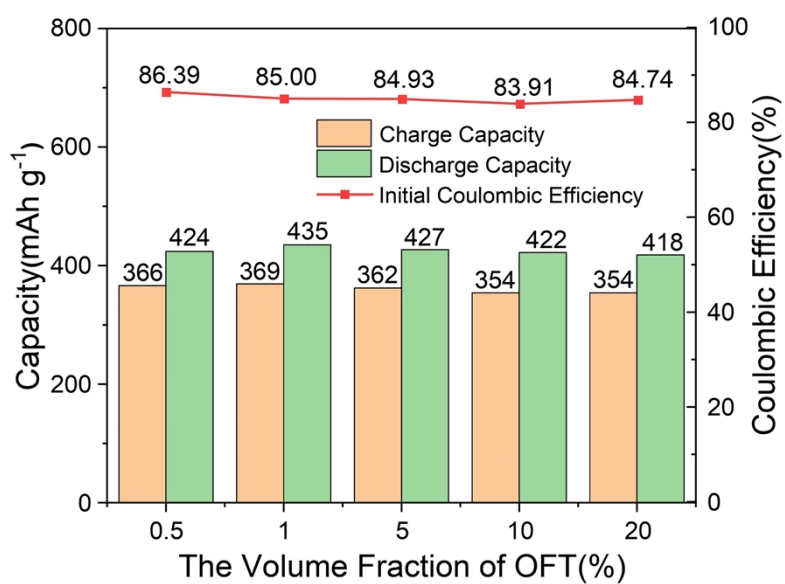
<sup>\*</sup>Corresponding authors. E-mail addresses: lih@whu.edu.cn.



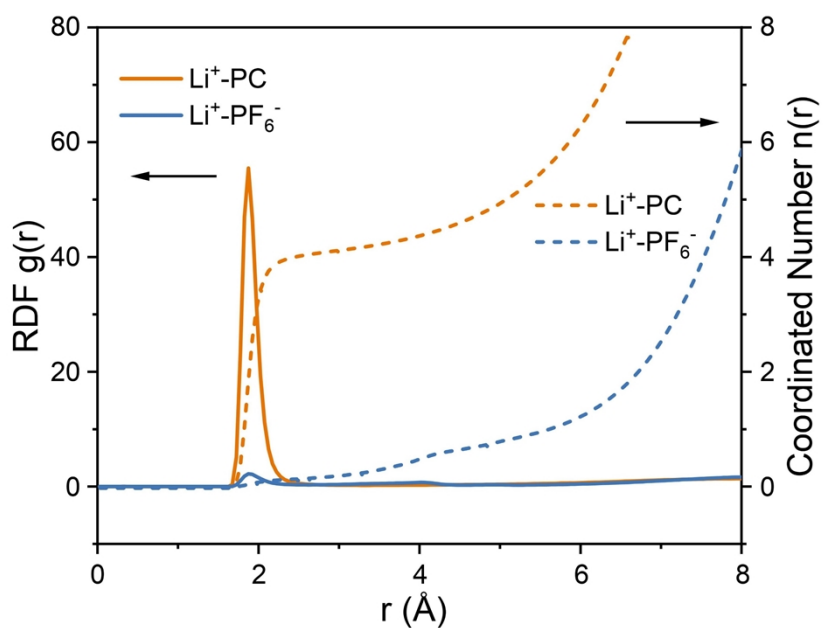
**Figure S1.** The highest occupied and lowest unoccupied molecular orbitals (HOMO and LUMO) of OFT and PC molecules.



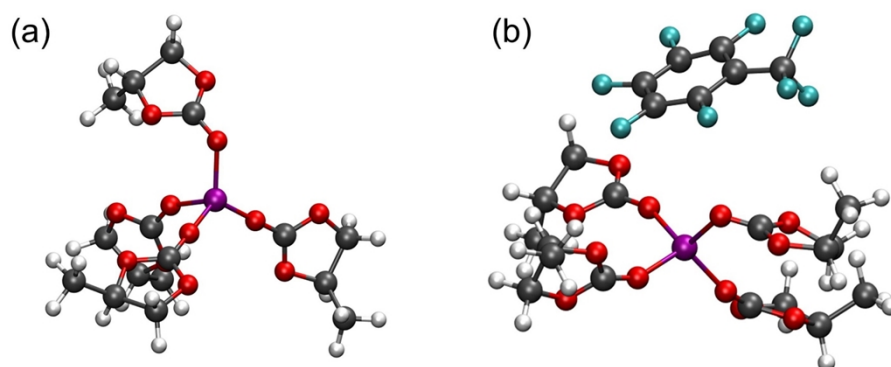
**Figure S2.** The initial discharge-charge curves of graphite electrode in 1 M LiPF<sub>6</sub>/PC electrolytes containing different amounts of OFT.



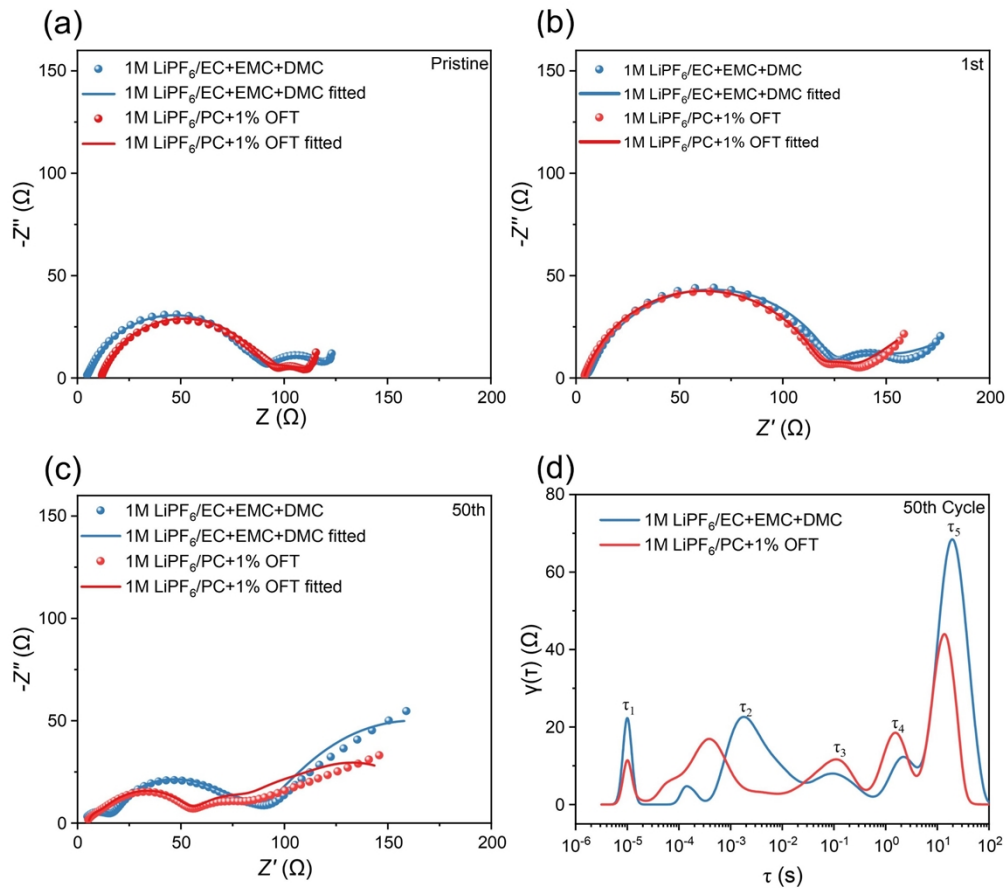
**Figure S3.** Comparison of initial Coulombic efficiency (ICE) and initial charge/discharge capacities of the graphite electrode in 1 M LiPF<sub>6</sub>/PC electrolytes containing different amounts of OFT.



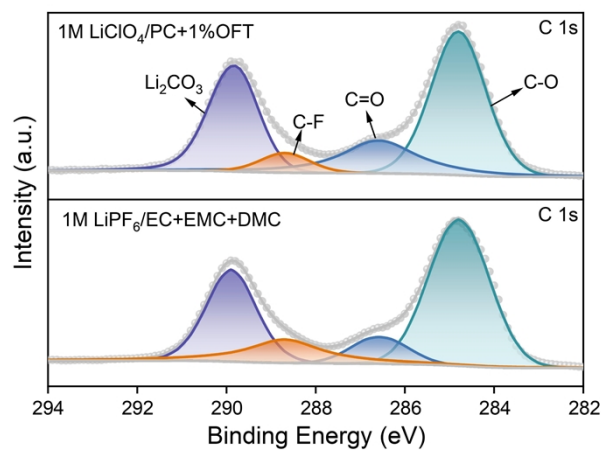
**Figure S4.** The radial distribution functions (RDFs) of solvent molecules at various distances from central  $\text{Li}^+$  and coordination numbers (CNs) of  $\text{Li}^+$  in 1 M  $\text{LiPF}_6/\text{PC}$  electrolytes.



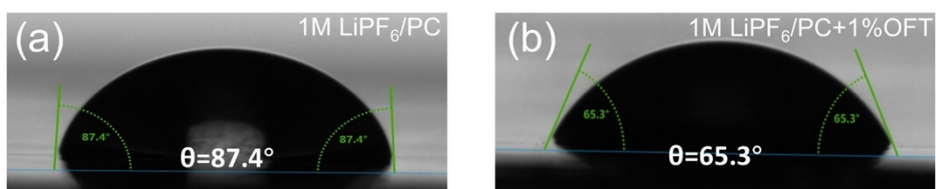
**Figure S5.** The optimized geometries of solvation structures of (a)  $\text{Li(PC)}_4^+$  and (b)  $\text{Li(PC)}_4^+$ -OFT.



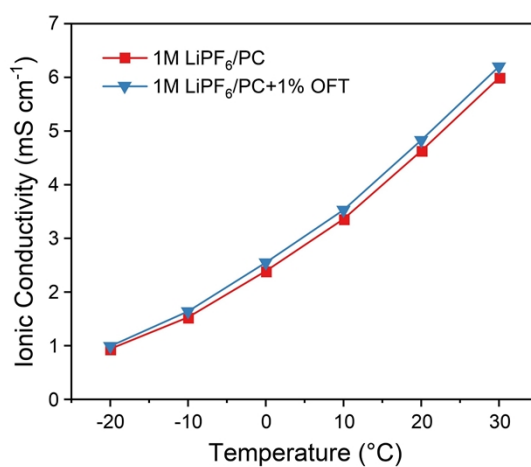
**Figure S6.** Electrochemical impedance spectra and their fitted curves for graphite electrode (a) before cycling, (b) after 1 cycle, (c) after 50 cycles and (d) DRT transformation for graphite electrode after 50 cycle.



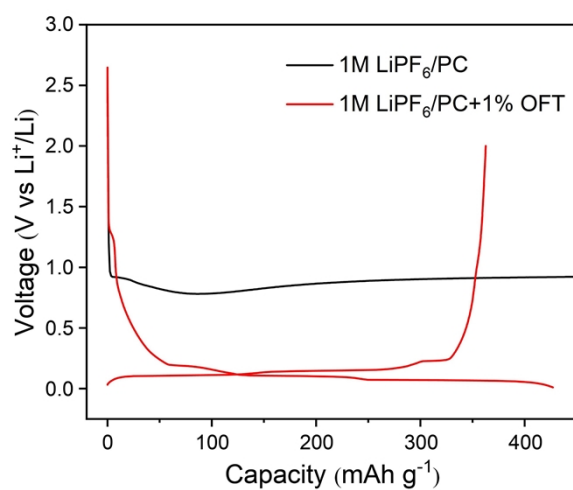
**Figure S7.** XPS C1s spectra of graphite electrode after cycling in 1 M LiClO<sub>4</sub>/PC+1% OFT and 1 M LiPF<sub>6</sub>/EC+EMC+DMC (1:1:1 by vol.) electrolyte.



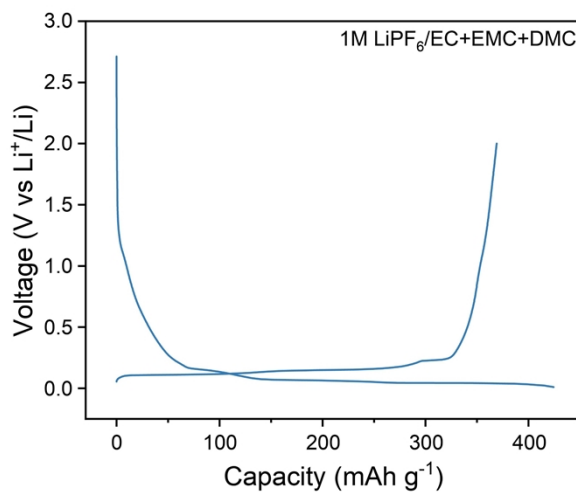
**Figure S8.** Electrolyte uptake of (a) 1M LiPF<sub>6</sub>/PC and (b) 1M LiPF<sub>6</sub>/PC+1% OFT electrolytes toward the PP separator.



**Figure S9.** Conductivities of 1M LiPF<sub>6</sub>/PC electrolytes with and without OFT at different temperatures.



**Figure S10.** The initial discharge-charge curves of graphite electrode in 1 M LiPF<sub>6</sub>/PC electrolytes with and without OFT.



**Figure S11.** The initial discharge-charge curves of graphite electrode in 1 M LiPF<sub>6</sub>/EC+EMC+DMC (1:1:1 by vol.) electrolyte.