Science Advances

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Supplementary Materials for

Understanding the molecular mechanism of pulse current charging for stable lithium-metal batteries

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Published 21 July 2017, *Sci. Adv.* **3**, e1701246 (2017)
DOI: 10.1126/sciadv.1701246

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SUPPLEMENTARY MATERIALS

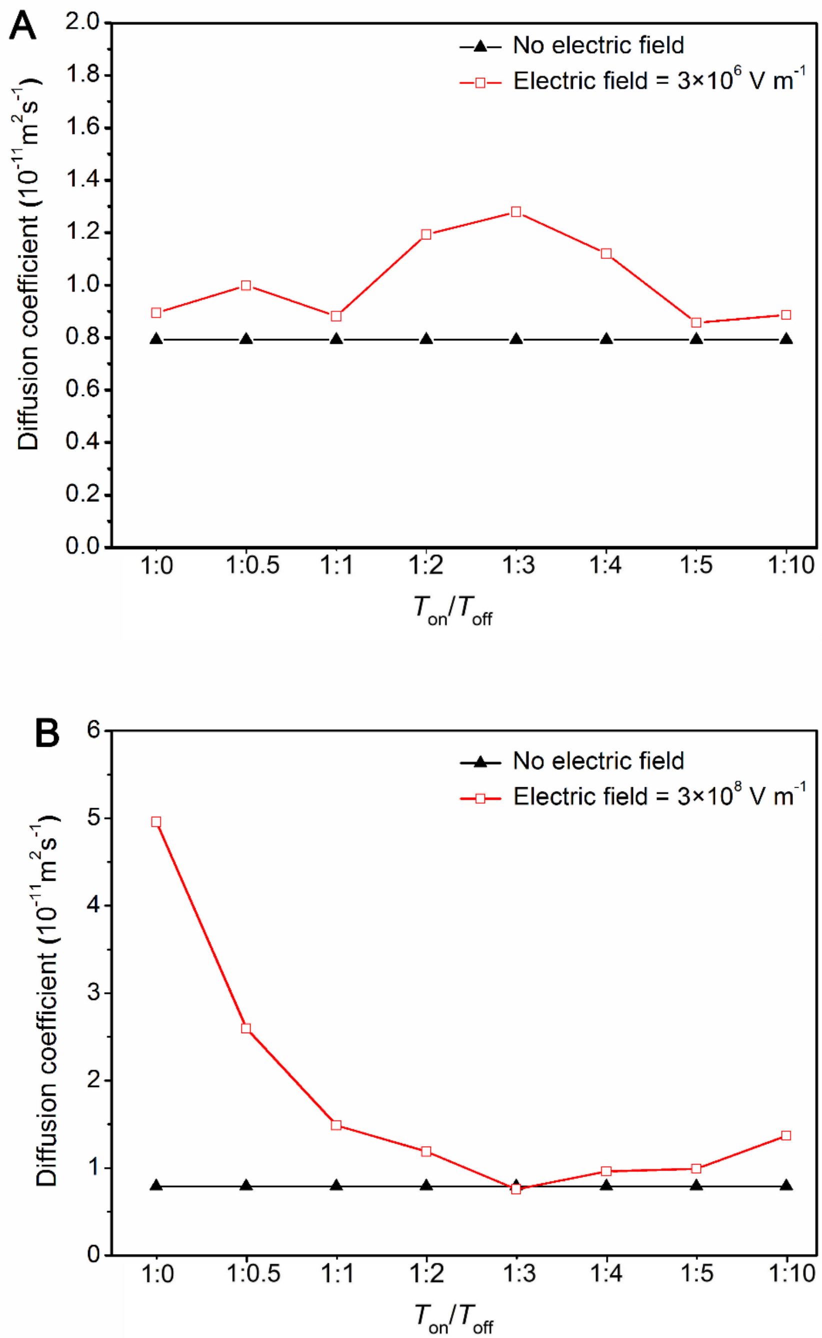

fig. S1. The impact of pulsed electric fields on the diffusivity of Li^+. (**A**) Diffusion coefficients of Li^+ in 1M LiTFSI/PC solutions under a static electric field with a strength of 3×10^6 V m^{-1}, under a pulsed electric field with the same strength, and with no electric field. (**B**) Diffusion coefficients of Li^+ in 1M LiTFSI/PC solutions under a static electric field with a strength of 3×10^8 V m^{-1}, under a pulsed electric field with the same strength, and with no electric field. The T_{on}/T_{off} ratios of pulsed electric fields are 1:0.5, 1:1, 1:2, 1:3, 1:4, 1:5, and 1:10. For convenience, a static electric field is shown as the one with a T_{on}/T_{off} ratio of 1:0. The diffusion coefficient of Li^+ is shown to be sensitive to the T_{on}/T_{off} ratio of the pulsed electric field with an optimal value at 1:3 in (A).

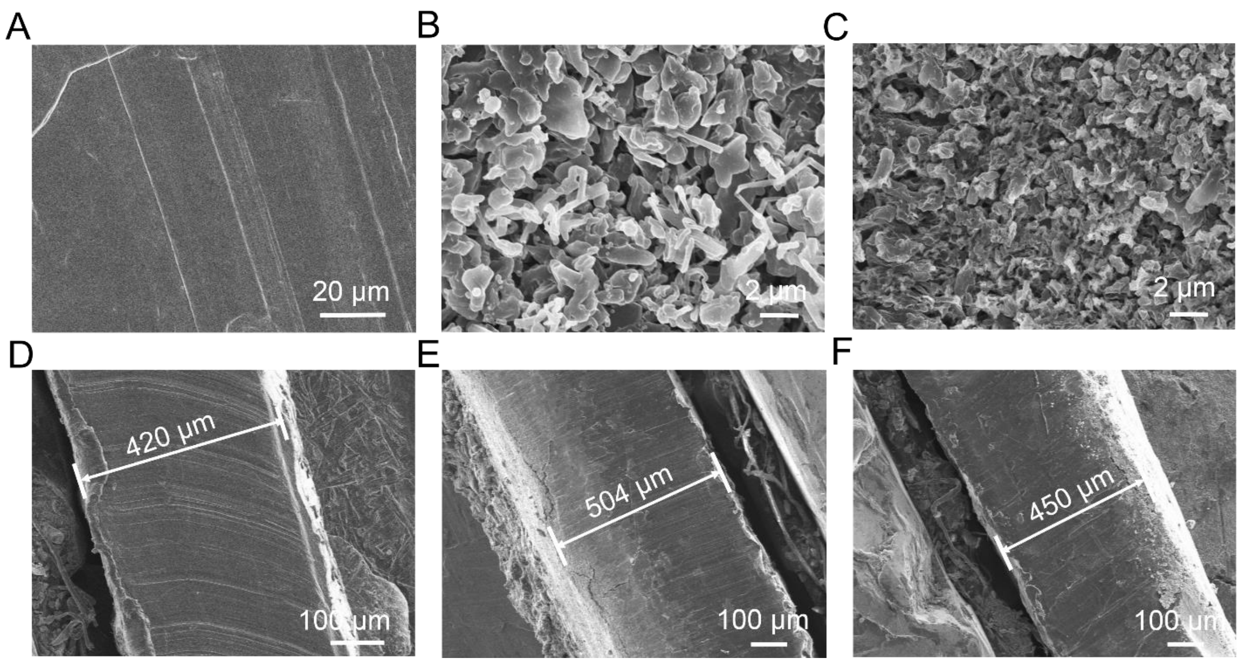

fig. S2. Surface and cross-section SEM images of Li-metal electrodes cycled with or without pulse current. Surface (**A**) and cross-section (**D**) SEM images of pristine Li-metal electrode. Surface (**B**) and cross-section (**E**) SEM images of Li-metal electrode from cells with constant current cycling. Surface (**C**) and cross-section (**F**) SEM images of Li-metal electrode from cells with pulse current cycling (T_{on}/T_{off} =1:5,T_{on}=1s). The electrodes were obtained from the Li/Li symmetrical cells before cycling and after being cycled for 8 times at current density of 3 mA cm^{-2} with a total capacity of 1mAh cm^{-2} for each half cycle.

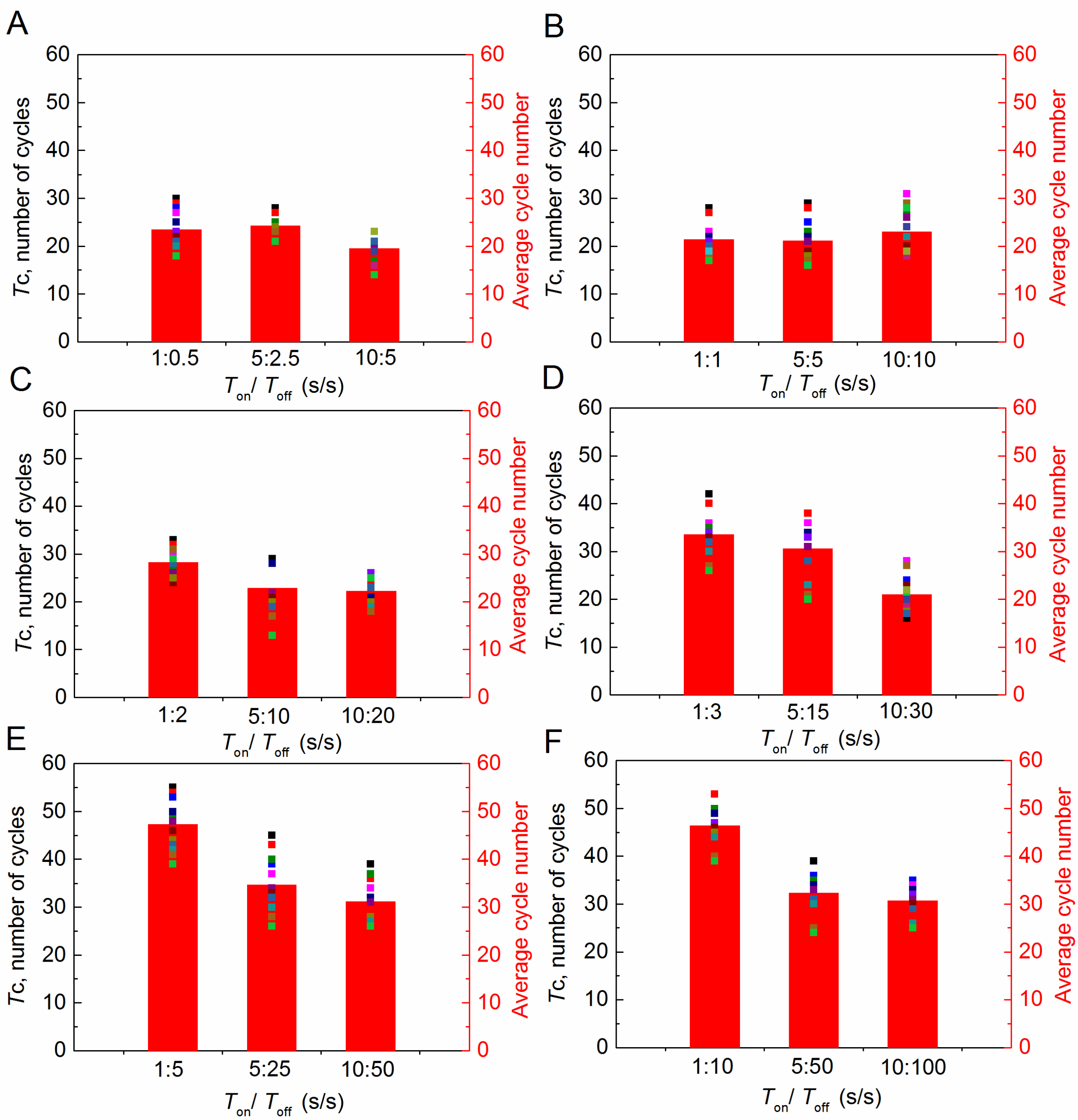

fig. S3. Cell lifetime at various T_{on} with fixed frequencies. Li/Li symmetrical cells were cycled with pulse current at fixed current density of 3 mA cm^{-2}. Cell lifetime at T_{on}/T_{off} = 1:0.5 (**A**), 1:1 (**B**), 1:2 (**C**), 1:3 (**D**), 1:5 (**E**), and 1:10 (**F**). Red columns show their average cell lifetime calculated from the measured 14 samples at each condition.

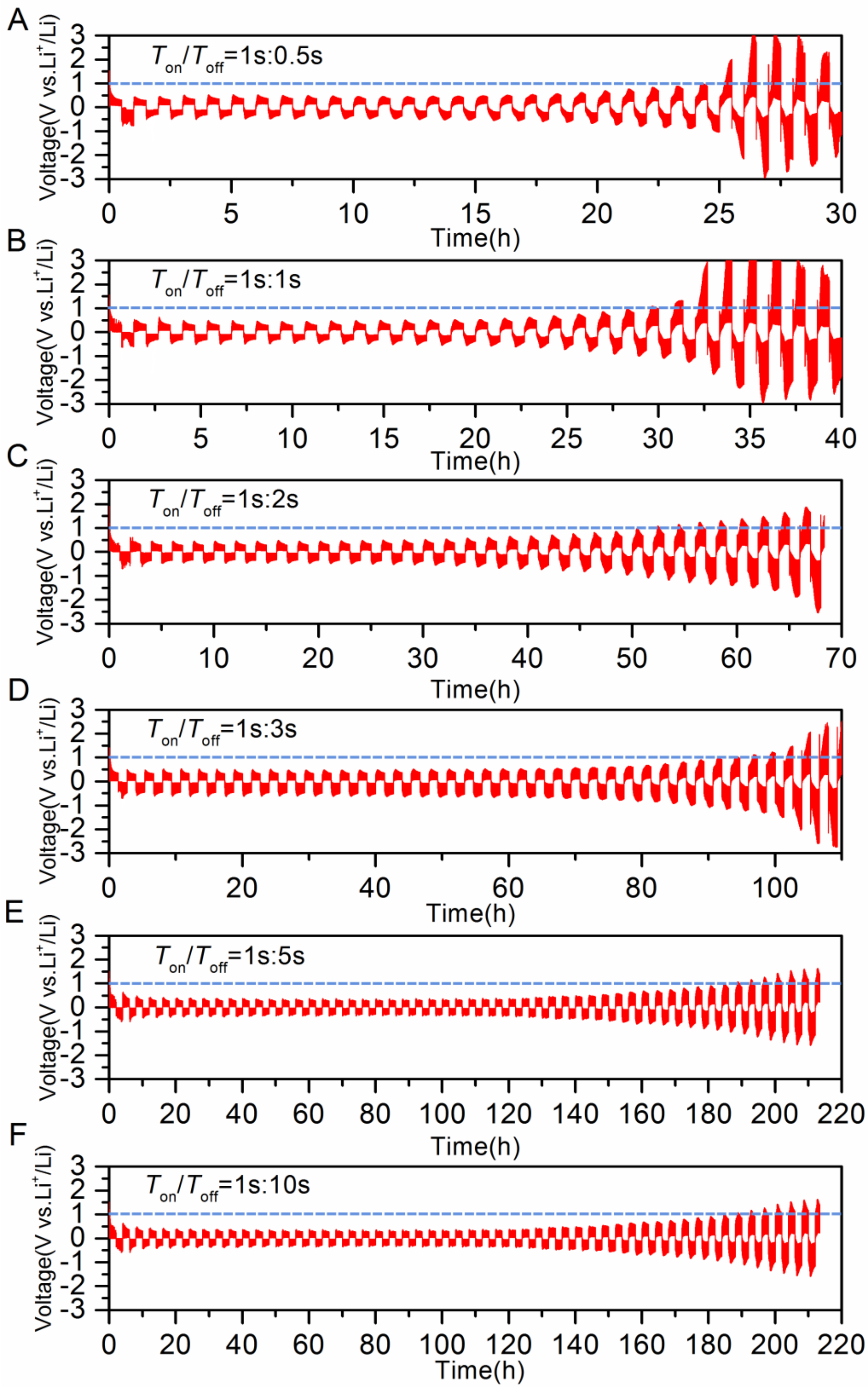

fig. S4. Voltage-time profiles of cells cycled with pulse current at fixed T_{on} = 1 s. Li/Li symmetrical cells were cycled at a fixed current density of 3 mA cm^{-2} at T_{on}/T_{off} = 1:0.5 (**A**), 1:1 (**B**), 1:2 (**C**), 1:3 (**D**), 1:5 (**E**), and 1:10 (**F**).

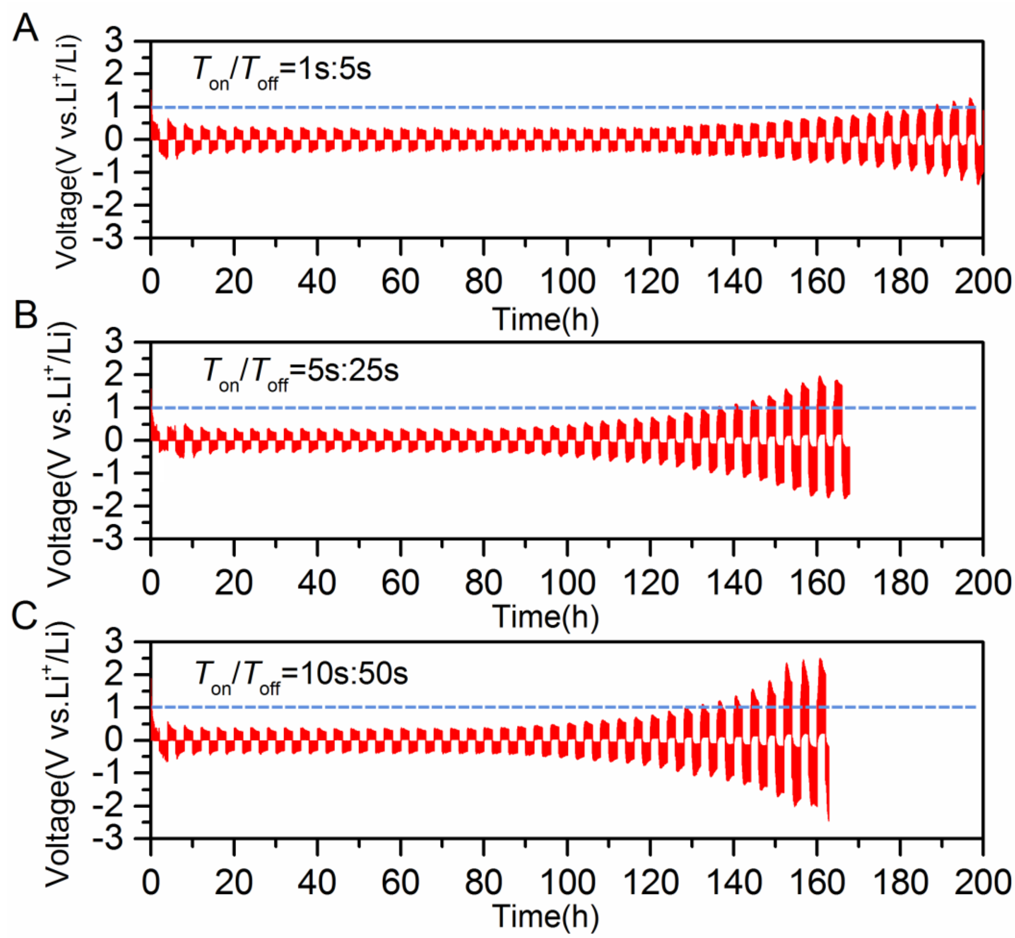

fig. S5. Voltage-time profiles of cells cycled with pulse current at fixed T_{on}/T_{off} = 1:5. Li/Li symmetrical cells were cycled with pulsed current at a fixed current density of 3 mA cm^{-2} at T_{on}= 1s (**A**), 5s (**B**), 10s (**C**).