Evaluating oxidation stability of redox mediator in lithium-oxygen batteries

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In lithium-oxygen (Li-O_2) batteries, employing organic redox mediators (ORMs) as mobile catalyst into the electrolyte system has been an essential strategy to lower the charging overpotentials Molecular designs of ORMs can also tailor their redox potential and rate of electron-transfer to improve the catalytic efficiency. However, the stability of ORMs in Li-O_2 cells was scarcely studied. In here, catalytic efficiency and stability of several important ORMs are assessed through in situ gas analysis and reactivity tests with singlet oxygen. Some well-known ORMs are detrimentally decomposed during the first cycle in Li-O_2 cells, whereas nitroxyl-radical-based ORMs bear the most stable and efficient response. Analogous nitroxyl-radical derivatives further increase round-trip energy efficiency and electron-transfer kinetics. This study also emphasizes the evaluation of chemical stability of ORMs, which are mandatory for the long-term cyclability in Li-O_2 cells.