Two-electron reduction of ethylene carbonate: a quantum chemistry re-examination of mechanisms

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Passivating solid-electrolyte interphase (SEI) films arising from electrolyte decomposition on low-voltage lithium ion battery anode surfaces are critical for battery operations. We review the recent theoretical literature on electrolyte decomposition and emphasize the modeling work on two-electron reduction of ethylene carbonate (EC, a key battery organic solvent). One of the two-electron pathways, which releases CO gas, is re-examined using simple quantum chemistry calculations. Excess electrons are shown to preferentially attack EC in the order (broken EC-) > (intact EC-) > EC. This confirms the viability of two electron processes and emphasizes that they need to be considered when interpreting SEI experiments. An estimate of the crossover between one- and two-electron regimes under a homogeneous reaction zone approximation, and a "kinetic phase diagram," are proposed.

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