

# Identifying degradation pathways and prognosis for lithium-ion batteries through a stochastic method

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This study identifies degradation pathways and characteristics of lithium-ion batteries through a multi-state Markov chain model and analyzes feasibility of the proposed method as a prognostic method for predicting the state of health. The proposed model would become a practical method for control- and design-enabling solution because the proposed method only requires measured capacity dataset to elucidate the degradation characteristics. Specifically, the proposed multi-state Markov chain model could determine the degradation state of a system by using conditional probabilities by using three phases of lithium ions: a sleeping phase, an active phase, and a dead phase. Lithium ions of each phase partially could convert to the other phase according to conditional probability and the number of cycles, determining the current capacity and state of LIBs. First, the sleeping phase represents potential lithium ions, which can be converted to an active phase that increases the capacity of LIBs. Second, the active phase describes the exact amount of lithium ions corresponding to the current capacity. Third, the dead phase denotes dead lithium ions, which can no longer be intercalated/deintercalated, decreasing capacity. Specifically, the proposed model comprises three sleeping states, two active states, and a dead phase. Each phase of the proposed model corresponds to the specific degradation modes and mechanisms including LAM, LLI, and CL. Stochastic parameters of the proposed model are estimated with two experimental degradation datasets, which one was originally conducted in author's laboratory<sup>1</sup>, and the other public degradation dataset, providing the conditional probability of each phase for accounting for degradation pathway dependency of LIBs. The estimated parameters of the proposed model could reveal that the conditional probability from the active phase to the dead phase increases according to the increase in temperature, inferring that high temperature accelerates a side reaction. Moreover, an effect of compressive force on degradation could be elucidated by using stochastic parameters of the proposed model<sup>2</sup>. Conditional probability from the sleeping state to the active state increase with optimal compression force, inferring that optimal force reduces contact loss<sup>3</sup>. Furthermore, this study demonstrates that the proposed model predicts the state of health only with partial data of the measured capacity, suggesting that the proposed method can be used as the prognostic method for the state of health<sup>4.5</sup>. Hence, quantitative comparison with other multi-state Markov chain models also shows that the proposed model outperforms other methods because the proposed model contains a non-homogeneous state and two sleeping states to account for the highly nonlinear and complex degradation characteristics of degradation for LIBs 6.7.8.9. The proposed model could accelerate the design optimization process and can become an effective prognostic method when capacity is only available.

### NOMENCLATURE

 $S_*(0) =$  initial capacity of each state  $P(S_a|S_b) =$  conditional probability from a to b LAM = loss of active material LLI = loss of lithium-ion inventory LIB = Lithium-Ion battery CL = Conductivity loss SOH = State of Health

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827

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