

Unraveling the Potassium Storage Mechanism in Graphite

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Potassium-intercalated Graphite intercalation compounds (K-GICs) are of particular physical and chemical interest due to their versatile structures and fascinating properties. Fundamental insights into the K^+ storage mechanism, and the complex kinetics/thermodynamics that control the reactions and structural rearrangements allow manipulating K-GICs with desired functionalities. Here *operando* studies including in-situ Raman mapping and in-situ XRD characterizations, in combination with density-functional theory simulations were carried out to correlate the real-time electrochemical K^+ intercalation/de-intercalation process with structure/component evolution. The experimental results, together with theoretical calculations, reveal the reversible K-GICs staging transition: $C \leftrightarrow \text{stage 5 (KC}_{60}) \leftrightarrow \text{stage 4 (KC}_{48}) \leftrightarrow \text{stage 3 (KC}_{36}) \leftrightarrow \text{stage 2 (KC}_{24}/\text{KC}_{16}) \leftrightarrow \text{stage 1 (KC}_8)$. Moreover, the staging transition are clearly visualized and an intermediate phase of stage 2 with the stoichiometric formula of KC_{16} is identified. The staging transition mechanism involving both intrastage transition from KC_{24} (stage 2) to KC_{16} (stage 2) and interstage transition is proposed. The present study promotes better fundamental understanding of K^+ storage behavior in graphite, develops a nondestructive technological basis for accurately capture non-uniformity in electrode phase evolution across the length scale of graphite domains, and offers guidance for efficient research in other GICs.