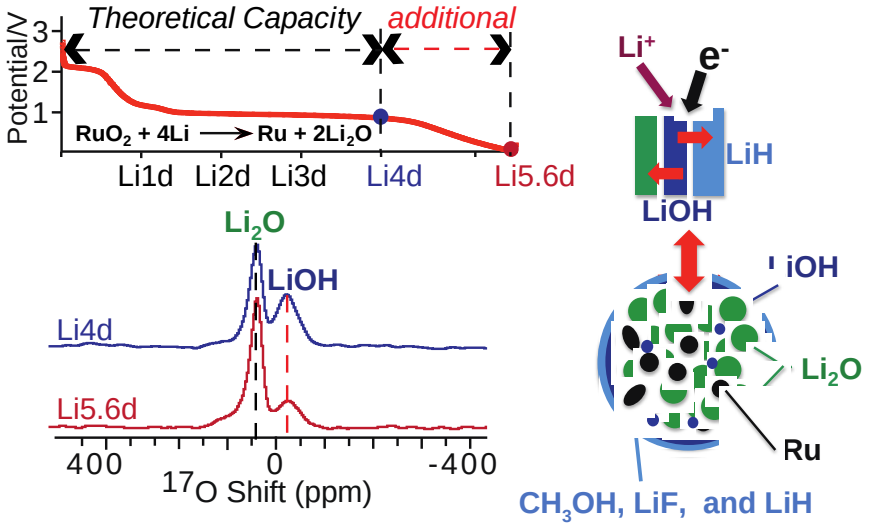
Origin of Additional Capacities Seen in Metal Oxide Lithium Ion Battery Electrodes

Scientific Achievement

A solid-state NMR protocol was demonstrated to study the buried and disordered solid-electrolyte-interphase (SEI). This method was applied to determine the cause of additional capacities found in metal oxide lithium ion battery conversion electrodes.

Significance and Impact

The study shows that the capacity observed in these systems beyond the theoretical value is due to electrolyte decomposition and LiOH formation and not a space charge mechanism, as claimed my some authors, helping to resolve a long-standing debate in the literature. The methodology developed as part of this study offers an effective way for investigating the nature of many phases present in the SEI, and is thus of wider interest (formation of a stable SEI is critical for the long term cyclability of all battery systems).



^{17}O NMR clearly shows the formation of Li_2O and LiOH on reaction with 4 Li (per RuO_2 formula unit; Li4d). Further reaction converts much of the LiOH to LiH and results in further SEI formation:

$LiOH + 2Li = LiH + Li_2O$ (Li5.6d)

Research Details

- ^{1}H, $^{6}Li/^{7}Li$, and ^{17}O high-resolution NMR spectra were acquired, allowing reliable identification and quantification of various phases formed in the SEI at different states of charge in the RuO_2/Li system
- Multidimensional NMR spectra were obtained to determine the relative spatial locations of SEI components.
- The method was applied to determine the sources of the additional capacities found in an RuO_2 electrodes.

Y.-Y. Hu, Z. Liu, K. W. Nam, O. J. Borkiewicz, J. Cheng, C. P. Grey *et al.* - Nat Mater (2013) in press (DOI: 10.1038/NMAT3784).

This work was performed at Stony Brook University, Cambridge University, Argonne National Lab, and Brookhaven National Lab.

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