Chemistry Colloquia

# Oxygen-redox Chemistry in High Energy Density Battery Cathodes


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**Moonstone** Seminar F

One of the biggest challenges facing lithium-ion batteries is how to increase their energy density. The cathode, typically a layered lithium transition metal oxide, represents a major limitation. One route to increase the energy density is to store charge at high voltage on the oxide ions in the cathode material. However, removing electrons from lattice oxide ions typically results in structural instability leading to voltage hysteresis and voltage fade over cycling. Understanding the mechanism behind oxygen redox is critical to overcoming these issues.

Our recent investigations into Li-rich cathodes have revealed that oxidized oxygen takes the form of $O_2$ molecules which are trapped in nanovoids in the structure. We have also shown that these trapped $O_2$ molecules can be reduced back to $O^{2-}$ on discharge providing a viable charge storage mechanism to explain oxygen redox. In this talk, I will discuss the evidence [1-3] for the formation and reduction of trapped $O_2$ and explore the impacts this has on the performance of oxygen redox cathodes, such as voltage hysteresis and fade [4,5]. I will show how the formation of $O_2$ extends to 4d and 5d transition metal oxides [5], disordered rocksalt cathodes [6] and even to non-Li-rich cathodes [7]. Finally, I will show that it is possible to suppress this structural change and undergo reversible, high voltage O-redox without voltage hysteresis [8]. Altogether, this understanding helps to explain the unusual properties of oxygen redox cathodes and informs how they might be harnessed to boost the energy density of batteries.


1. House, R. A. et al. ***Nature*** 577, 502-508 (2020).
2. House, R. A. et al. ***Nature Energy*** 8, 777–785 (2020).
3. House, R. A. et al. ***Energy & Environmental Science*** 15, 1 376-386 (2022).
4. House, R. A. et al. ***Nature Energy*** 6, 781-789 (2021).
5. Marie, J. J. et al. ***Nature Materials*** 23, 818–825 (2024).
5. House, R. A. et al. ***Nature Communications*** 12, 2975 (2021).
6. McColl, K. et al. ***Nature Communications*** 13, 5275 (2022).
7. Juelsholt, M. et al. ***Energy & Environemental Science***, 17, 2530-2540 (2024).
8. House, R. A. et al. ***Nature Energy*** 8, 351-360 (2023).


**Robert House** is an Associate Professor in the Department of Materials, Fellow of ZERO Institute at Oxford and Non-Tutorial Fellow of Keble College. He holds a Research Fellowship with the Royal Academy of Engineering and affiliations with the Faraday Institution and Henry Royce Institute for advanced materials.

Professor House's research interests are in novel sustainable energy materials, particularly for electrochemical energy storage, and in developing and applying advanced X-ray, neutron and muon scattering techniques to study ion diffusion, charge storage mechanisms and structure-function relationships in energy storage materials.

His research group are actively engaged in materials discovery and development for next generation batteries, and work closely with a range of industry partners. Their activities cover future lithium-ion technology, and more sustainable alternative chemistries such as sodium-ion and magnesium-ion.