Understanding heterogeneous transformations in Li-S batteries through *operando* diffraction mapping

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While most *operando* studies of battery materials during electrochemical cycling are carried out at a single point under the assumption of heterogeneity, this approach is inadequate for Li-S batteries which we have found to have 1 – 2 orders of magnitude more heterogeneity than classic layered battery systems. Comparative studies of the phase evolution during cycling were carried out at many different points on a series of pouch cell batteries and the collective information obtained from the many thousands of diffraction patterns collected during these studies provides clear mechanistic information that contradicts prior conclusions obtained from studies done at a single point. Furthermore, novel *operando* cells with large windows (> 25 cm^2) have been designed and deployed to map the entire area of pouch cell batteries under isostatic pressure to test the ability of pressure to mitigate the heterogeneity in Li-S batteries. Our experiments provide clear evidence of the central role that the electrolyte plays in the initial and time-dependent heterogeneity of Li-S pouch cell batteries and highlight the challenges that will need to be overcome to realize high-energy density Li-S batteries with long lifetimes.

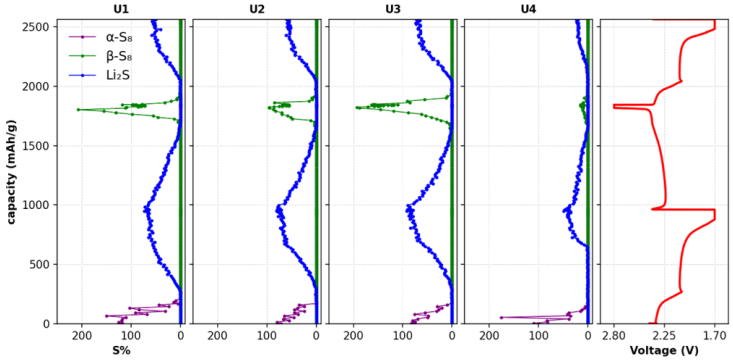


Figure 1. *Position-dependent speciation during electrochemical cycling of a Li-S pouch cell battery resolved from synchrotron operando powder diffraction studies.*

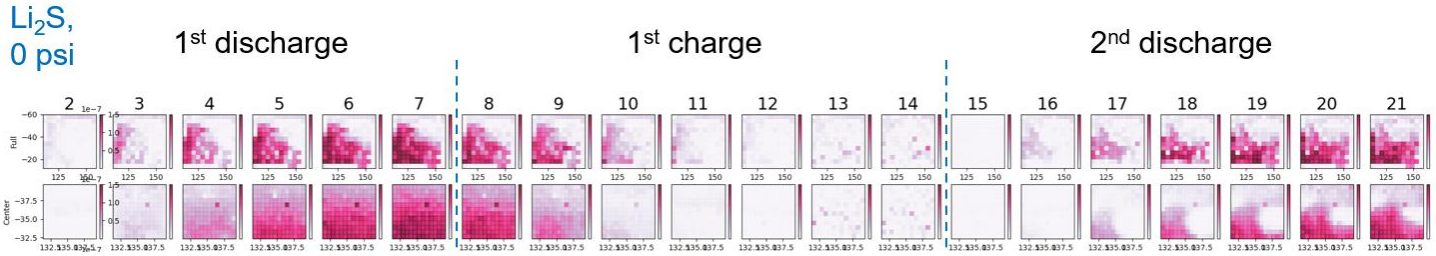


Figure 2. *Evolution of Li_2S abundance during Li-S pouch cell battery cycling determined from operando powder diffraction maps collected across the full pouch cell area (top row) and in a small region at the center of the cell (bottom row). The strong spatial heterogeneity is pressure-dependent and the length scale of features highlights the electrolyte influence.*