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Development of operando solid-state Nuclear Magnetic Resonance Spectroscopy (ssNMR) for metal-oxygen batteries

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Rechargeable metal-O$_2$ batteries have attracted much attention in recent years as a possible alternative to the widely used lithium-ion batteries. This is particularly the case for lithium and sodium-oxygen batteries, due to their potential high energy density.$^1$ However, great challenges remain in the development of metal-O$_2$ batteries, including the stabilization of the discharge products, poor cyclability and the need for new cathode design. On a more fundamental aspect, one much completely grasps the understanding of the underlying electrochemical mechanisms taking placed inside the M-O$_2$ battery. Clear identifications of the discharge electrochemical pathways and their products (M-O$_2$, M$_2$O$_2$,…), as well as the reactivity of the electrolyte, are crucial. Discharge products have already been observed using many ex situ techniques such as XRD or Raman, however it may not provide a realistic view of the reactions and may lead to biased interpretation. Conversely, In situ/operando techniques$^2$ can capture the dynamic changes unequivocally in the cell environment during cycling and provide unprecedented information on intermediate phases that may be undetectable through ex situ analyses.

In the past few years, ex situ ssNMR emerged as a valuable tool for characterizing metal-O$_2$ cells on ex situ discharged and charged samples (Fig 4).$^3,4$ Operando ssNMR appears thus of considerable interest to monitor the structural and electronic changes in M-O$_2$ upon cycling and to obtain valuable insights on the transition of chemical phases. To this aim, we recently designed an operando cell dedicated to metal-O$_2$ batteries that enables for O$_2$ gas to react at the cathode without compromising the NMR experiments. The electrochemical performance in the Li-O$_2$ configuration is quite similar to what is expected in the literature and validates the design of the cell. We will detail the working principle of the cell, its performance, and present the evolution of NMR signals during cycling.

Operando $^7$Li NMR on a Li-O$_2$ cell
References