New approaches for studying structure and dynamics in electrodes for lithium and beyond lithium batteries

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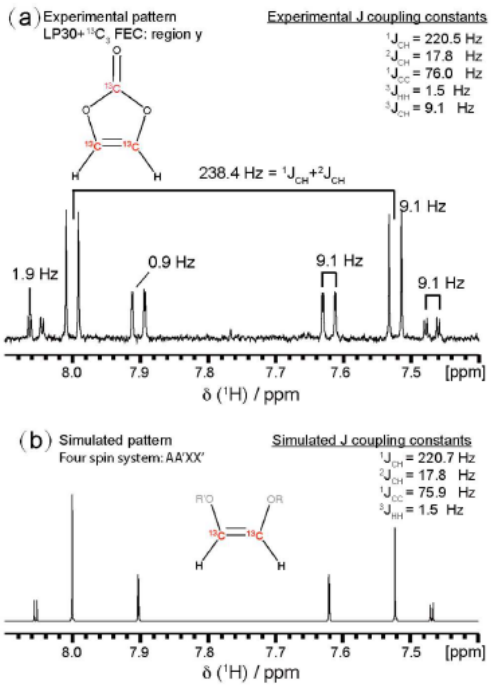

This talk will describe some recent advances in the use of NMR spectroscopy and related approaches to study structure and dynamics of both the bulk electrodes and the interface (interphase) between different components.

SEI Chemistry: The primary aim of this study was to understand how the electrolyte additive fluoroethylene carbonate (FEC) enhances the stability of the solid electrolyte interphase (SEI) formed on silicon anodes. Key developments include the use of ^{13}C enriched FEC, which provided compelling evidence (via the use of ^1H and ^{13}C 1 and 2D NMR spectroscopy) for the defluorination of FEC to form soluble vinoxyl species ($HCOCH_2OR$) and VC. The formation of VC, rather than LVDC, was unambiguously determined by using the J coupling pattern that arises as a result of the uniform ^{13}C labelling of FEC and the subsequent breakdown products (Fig 1). Oligomers with characteristic peaks due to vinoxyl species were also identified, which presumably react further to form insoluble polymeric species in the SEI, with similar cross-linking groups. ^{29}Si MAS NMR and dynamic nuclear polarization (DNP), the latter allowing experiments on non-enriched materials to be performed, were used to perform a detailed study of the Si-organic interface and its change on cycling.

Structure solution and dynamics: The use of a combination of NMR, theory and pair distribution function (PDF) analysis methods to solve amorphous and disordered anode structures will be outlined, focussing on Na anode materials, including C and Sn. The use of pulse field gradient (PFG) methods to study transport in electrode materials and to correlate this with rate studies will be outlined for a series of electrodes and electrode structures. Finally, the use of magnetic resonance imaging (MRI) to investigate dendrite formation will be discussed.

References:

1. Jin, Y.; Kneusels, N.-J. H.; Magusin, P. C. M. M.; Kim, G.; Castillo-Martinez, E.; Marbella, L. E.; Kerber, R. N.; Howe, D. J.; Paul, S.; Liu, T.; Grey, C. P. *J. Am. Chem. Soc.*, **2017**, *139*, 14992.