

Lithium iron methylene diphosphonate, a new organic-inorganic hybrid positive electrode material for Li-ion batteries

Sebastian Schmidt¹, Denis Sheptyakov², Claire Villevieille¹, Petr Novák¹, and Sébastien Sallard¹

¹ Paul Scherrer Institut, Electrochemistry Laboratory, CH-5232 Villigen PSI, Switzerland

² Paul Scherrer Institut, Laboratory for Neutron Scattering and Imaging, CH-5232 Villigen PSI, Switzerland

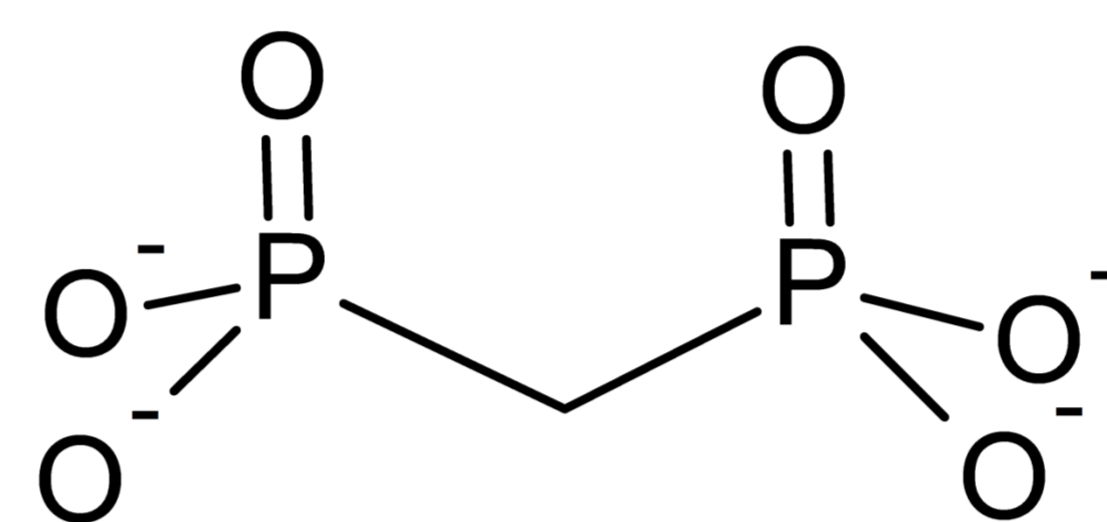
sebastien.sallard@psi.ch



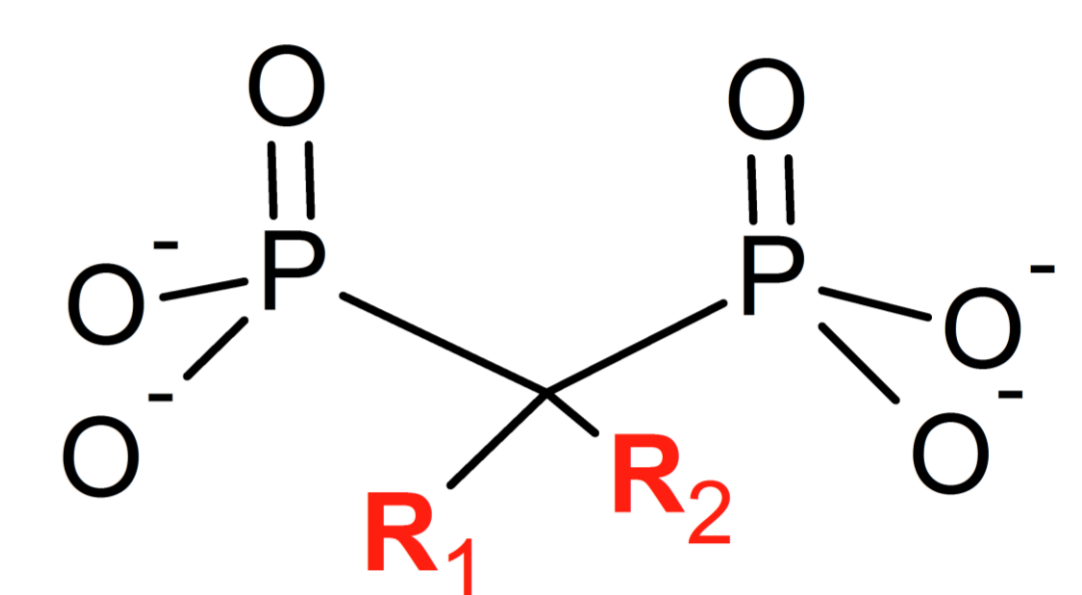
Methylene diphosphonate

- Ligand for organic-inorganic hybrid materials
- Alternative to carboxylic acid groups as ligands as used in other hybrid battery materials
- Varying substituents (R_1 , R_2) → quasi-infinite possibilities to design new materials

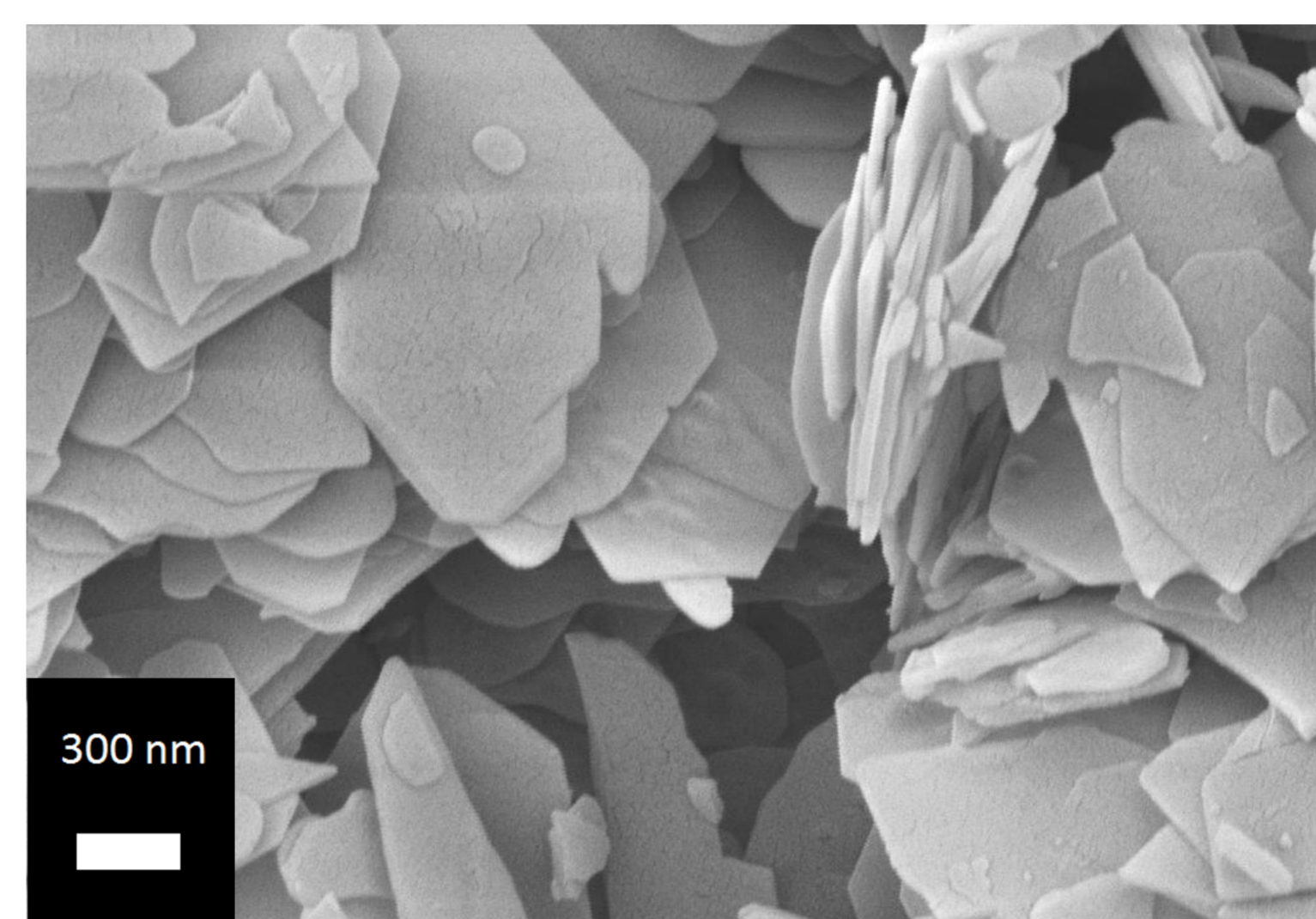
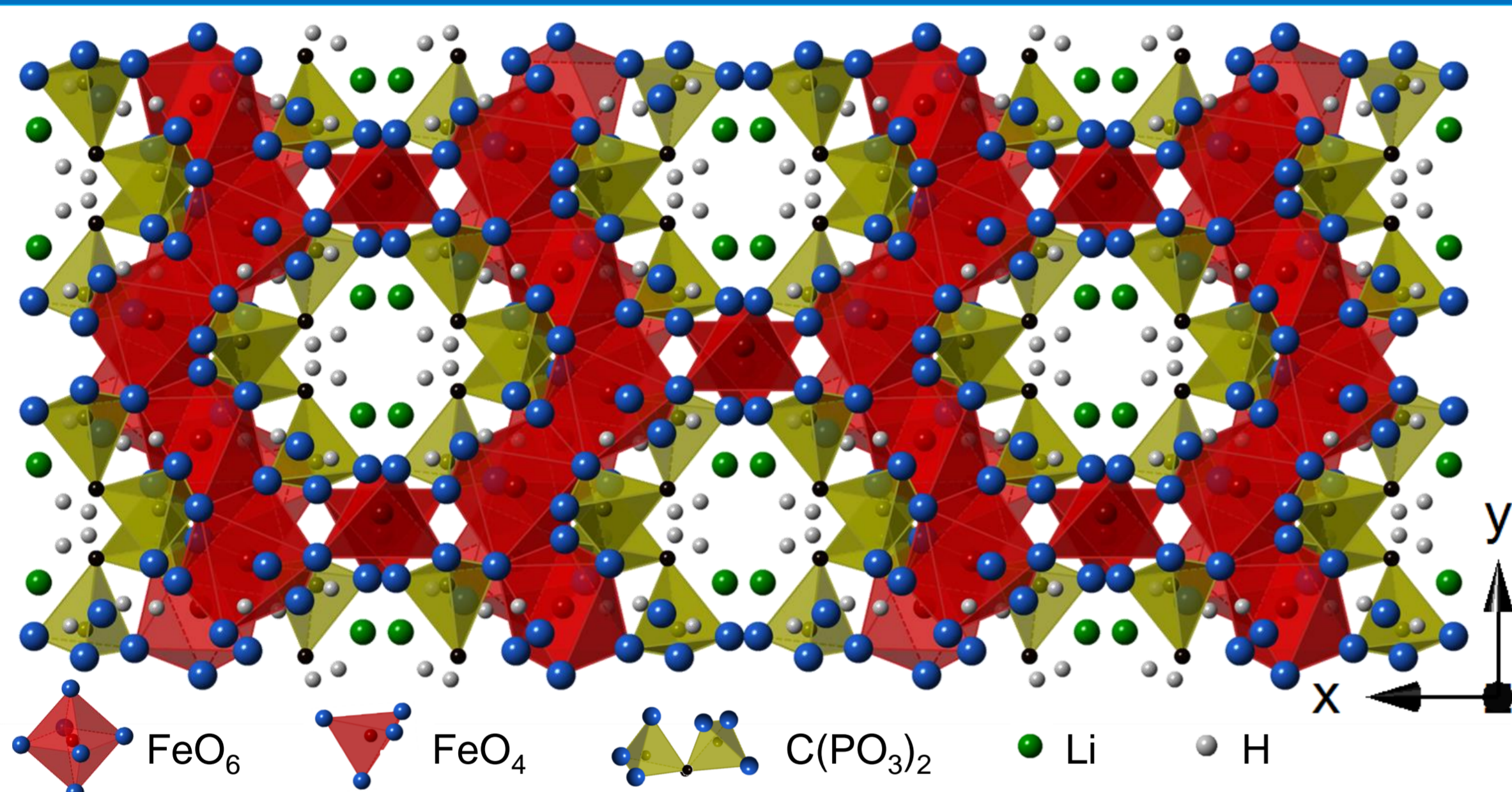
Methylene diphosphonate:



Perspective:



Structure & morphology



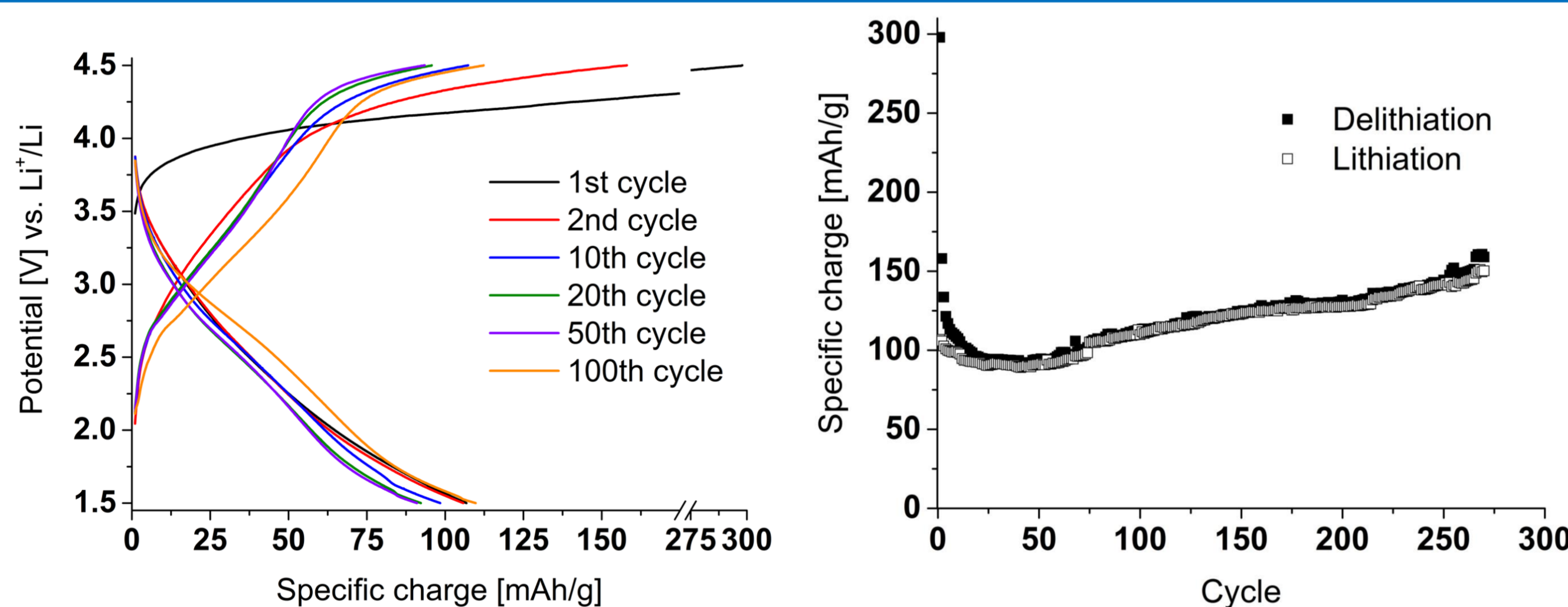
Refined structure:

- Monoclinic space group
- Octahedrally and tetrahedrally coordinated Fe sites
- Li sites located in channels

SEM:

- Sub-micrometric platelets, 40 – 50 nm thickness

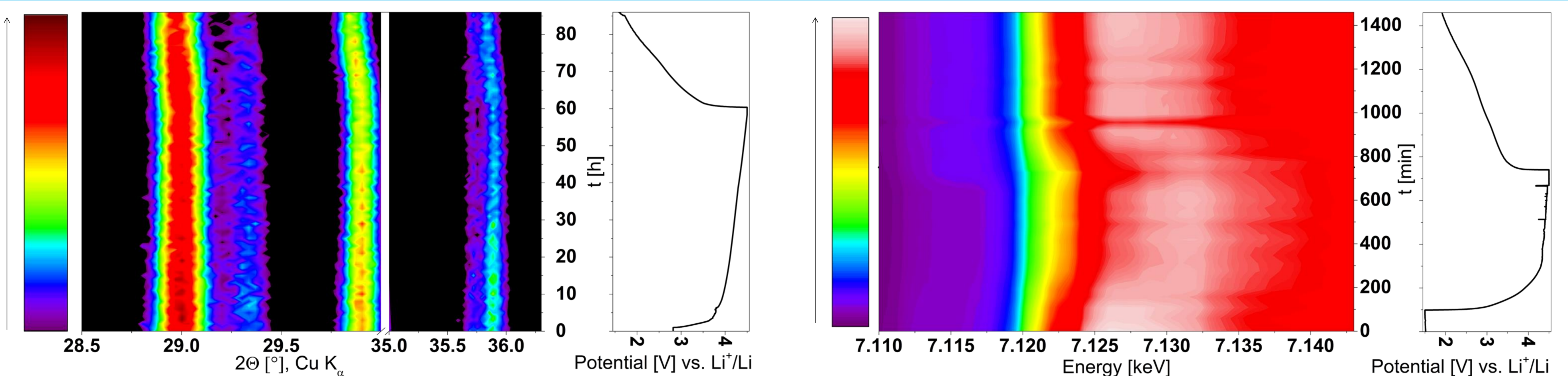
Electrochemical properties



- Increase in specific charge after 60 cycles, slight change in galvanostatic profile → suspected change in morphology [1].
- Ex situ IR (not shown) → diphosphonate still present after 40 cycles.

[1] Wang et al., *Advanced Energy Materials*, 2013, 3, 606 – 614.

Operando XRD / operando XANES



XRD: Only small change in the unit cell parameters

XANES: Fe(II) ↔ Fe(III) reversible cycling

Acknowledgements

The authors would like to express their sincere gratitude to:

- The Swiss National Science Foundation (SNF) for funding (project no. 200021_146224)
- The microXAS beamline and the MS powder beamline teams at the Swiss Light Source (SLS) for the experimental support

Conclusions & Outlook

- ✓ Lithium iron methylene diphosphonate can be cycled in half-cells
- ✓ Channels allow Li insertion/extrusion without strong disturbance of the unit cell parameters
- Investigations on the ligand and on the interfaces
- New materials with other metal and/or diphosphonate