Oxysulfide Solid Electrolytes for Lithium- and Sodium-Ion All-Solid-State Batteries

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An important milestone in the development of bulk all-solid-state batteries has been the discovery of thio-LISICON and related materials by Kanno et al. [1]. A key aspect in that discovery was the replacement of small, hard oxide ions in the LISICON structure with larger, softer sulfide ions, resulting in solid crystalline materials that can conduct Li^+ ions faster than liquid electrolytes [2]. Sulfide superionic conductors also present advantages in processing due to their mechanically compliant nature. However, sulfides have been shown to be quite unstable in the operating voltage range of batteries, and decompose in contact with electrode materials, limiting battery performance [3]. On the other hand, the original oxide materials are quite stable in wide voltage ranges. As such, we investigate oxysulfide compositions in search of materials that can satisfy the stringent requirements for application in an all-solid-state battery in terms of conductivity, stability and processability.

We follow a synergistic approach of synthesis, characterisation and modelling to elucidate the fundamental effects of composition and structure on the above-mentioned properties. Preliminary studies are performed on well-studied model systems composed of orthophosphate tetrahedra ($[PO_4]^{3-}$ and $[PS_4]^{3-}$). Their bulk and local structures are investigated using neutron and x-ray diffraction, vibrational spectroscopy and nuclear magnetic resonance (NMR). Ionic conductivity is investigated over multiple scales using electrochemical impedance spectroscopy, NMR, and large-scale, force-field molecular dynamics simulations [4], [5]. (Electro-)chemical stability is investigated using electrochemical cycling, thermal- and structural analysis techniques.

This multidisciplinary approach allows us to screen and isolate solid electrolyte materials for all-solid-state battery applications, prepared through tailored synthesis routes.

References:

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