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Quasi-Elastic Neutron Scattering Studies on Solid Electrolytes for solid state Lithium Batteries

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The development of better batteries is paramount for market penetration of electrical vehicle and integration of renewable energy sources into the grid. For today's best technology, state-of-the-art lithium-ion battery, limited improvement in capacity and cost are expected because of the use of organic liquid or gel electrolytes limiting the choice of electrode materials and exposing to safety concerns. A choice alternative is to use solid electrolytes instead.

The high temperature phase of lithium borohydride (LiBH_4), hexagonal crystal structure at temperature above 383 K, is a fast Li^+ conductor ($\sigma \sim 10^{-4} \text{ } \Omega \cdot \text{cm}^{-1}$) [1]. This property is kept at room temperature by stabilizing the hexagonal phase with Li halides [2] or confining LiBH_4 in nanoporous scaffolds.[3]

We have studied the Li^+ diffusion via Density Functional Theory (DFT) coupled to Quasi-Elastic Neutron Scattering (QENS). DFT shows that lithium defects such as Frenkel pairs are easily formed at room temperature ($E_f = 0.44 \text{ eV}$) and low energy barriers (0.3 eV) are found between stable defect sites, giving rise to high defect mobility. The most favorable mechanism for the Li^+ conduction is calculated to occur in the hexagonal plane. The QENS results at 380 K show long range diffusion of Li^+ , with jump lengths of one unit cell in the hexagonal plan and jump rates in agreement with DFT results. At 300 K, the data reveal jumps of shorter length ($\sim 2 \text{ } \text{Å}$), which could correspond to jump of Li^+ interstitials to intermediate lattice sites, in agreement with DFT.[4]

QENS was also used to probe the dynamic of the BH_4^- anions in composite electrolytes made of LiBH_4 and SiO_2 aerogels. Four quasi-elastic components were found in two different temperature domains. At low temperature, $< 170 \text{ K}$, the components are an order of magnitude broader than those at high temperature, however still below the phase transition. The narrower components are associated with reorienting BH_4^- anions in crystalline LiBH_4 , while the broader components, with much more rapidly reorienting BH_4^- , can be associated with the fraction of LiBH_4 located at the SiO_2 surfaces, reflecting a disruption of the bulk crystal lattice. It suggests that the high Li^+ conductivity takes place at the interface between LiBH_4 and SiO_2 .

Solid-state Lithium-Sulfur batteries, built around $\text{LiBH}_4/\text{SiO}_2$ solid electrolytes show good cyclability and almost full usage of the cathode capacity ($\sim 1500 \text{ mAh/g}$).[5]

References

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