

Rapid Li Spin Dynamics in the Interfacial Regions of the Nanocrystalline Solid Electrolyte LiBH₄

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Currently, all-solid-state batteries with ceramic electrolytes and lithium metal anodes, for example, represent an attractive alternative to conventional Li-ion batteries. So far many materials have been reported as promising candidates for such systems.^{1,2} Their dynamic properties and electrochemical stabilities are feverishly studied to identify those materials that may indeed replace flammable liquid electrolytes.

LiBH₄, especially when available in a nanocrystalline form, belongs to one of these promising solid electrolytes.² Besides its good compatibility with metallic lithium it shows a high ionic conductivity above 113 °C making it a suitable candidate for high-temperature solid-state batteries.³ In general, nanostructured ceramics, if prepared by high-energy balling from the coarse-grained counterparts, are composed of crystalline grains surrounded by structurally disordered interfacial regions. These areas are considered to be responsible for increased ion dynamics in these materials.

The aim in this work was to separately study the diffusivity of the fast Li ions in the large volume fraction of interfacial regions from an atomic scale point of view. Therefore, we used nuclear magnetic resonance (NMR) to record purely diffusion-induced ⁷Li NMR spin-lattice relaxation magnetization curves. Indeed, the transients are composed of two components. We attribute the fast relaxing component to the highly mobile Li ions in the interfacial regions. These ions showed much lower activation energies than those in the crystalline grains. NMR line shapes, recorded under static conditions, corroborated this point of view.⁴

References:

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