

Mechanosynthesis of Solid Electrolytes – Preparation, Characterization and Li Ion Transport Properties of Garnet-type Al-doped $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ Crystallizing with Cubic Symmetry

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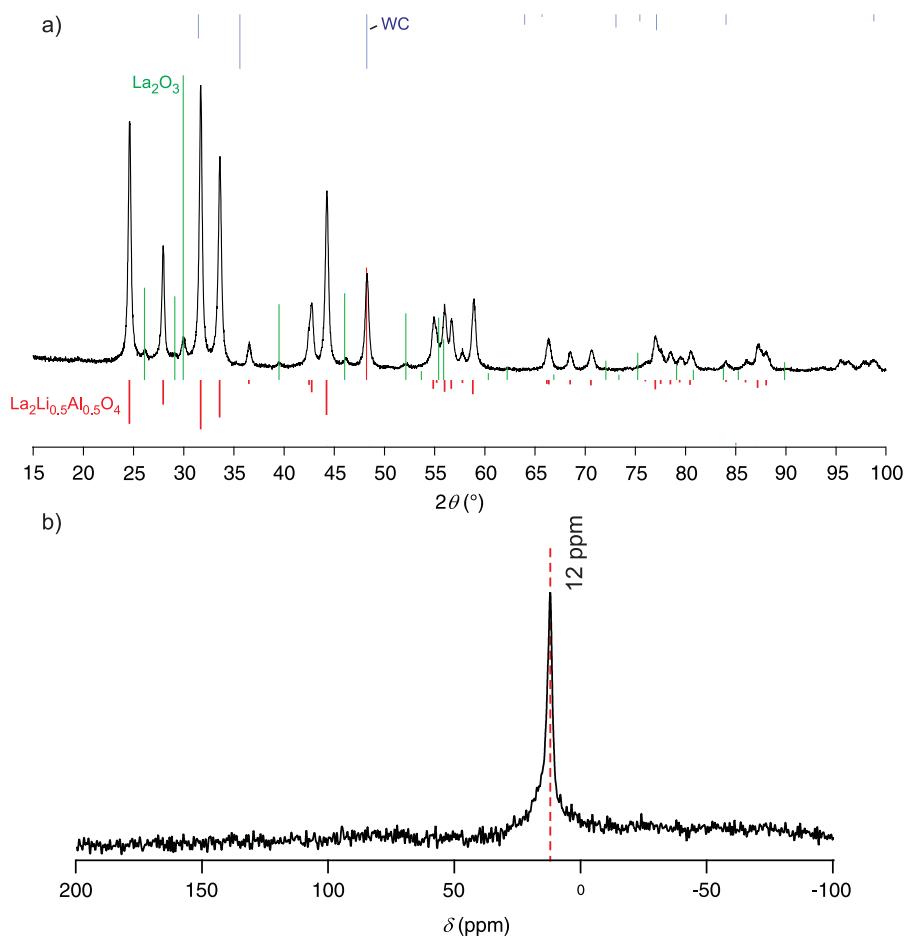


Fig. S1: a) XRPD pattern of mechanosynthesized $\text{La}_2\text{Li}_{0.5}\text{Al}_{0.5}\text{O}_4$ (8 h milling, 6 h annealing at 873 K (JCPDS 40-1167)). As an impurity phase a small amount of La_2O_3 (JCPDS 83-1344) can be observed. b) the corresponding ^{27}Al MAS NMR spectrum (recorded at 14.1 T and a spinning speed of 60 kHz with a 1.3 mm MAS probe (Bruker)) consisting of a single NMR line with a chemical shift of 12 ppm.

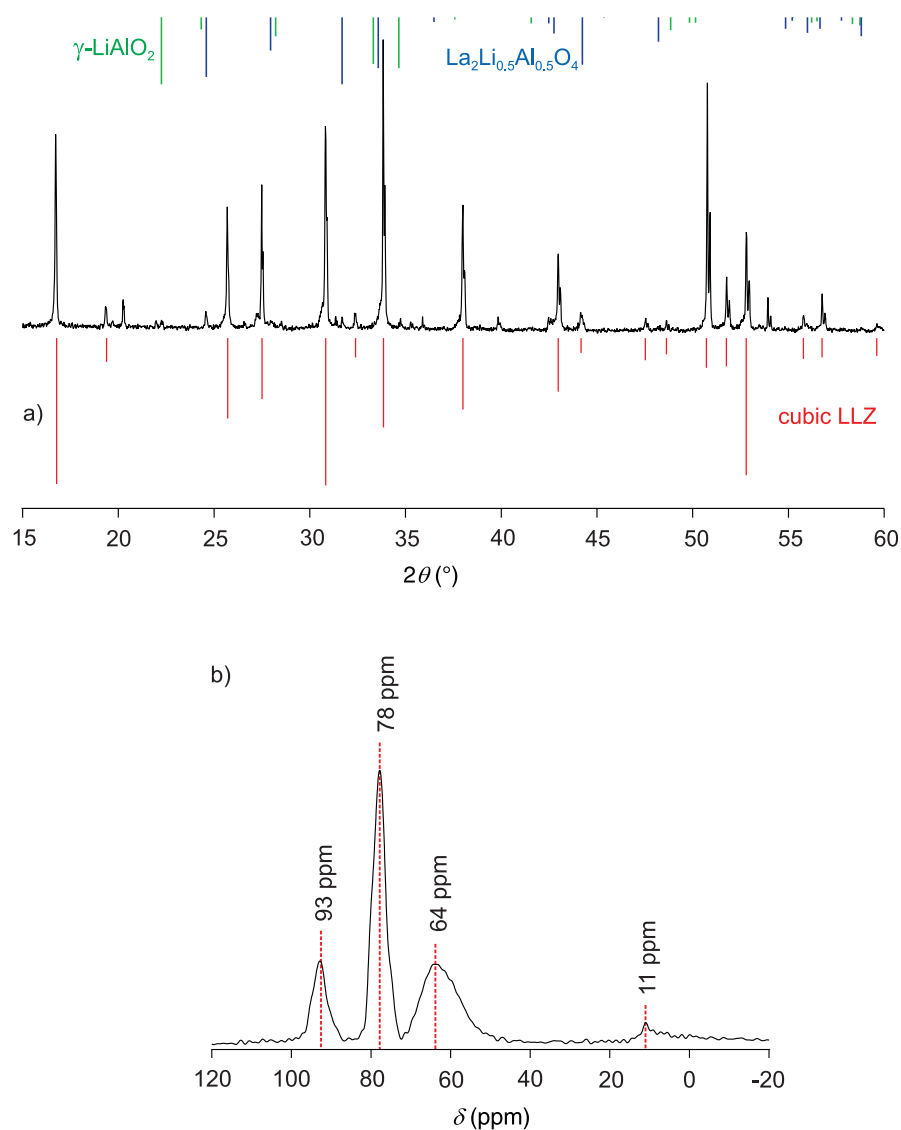


Fig. S2: a) XRPD pattern of a sample with a nominal composition of $\text{Li}_{7.75}\text{Al}_{0.40}\text{La}_2\text{Zr}_2\text{O}_{12}$ after annealing at 1500 K for 15 h. Several impurity phases like $\text{La}_2\text{Li}_{0.5}\text{Al}_{0.5}\text{O}_4$, γ -LiAlO₂ and unknown phases can be observed. b) the corresponding ²⁷Al MAS NMR spectrum (recorded at 14.1 T and a spinning speed of 30 kHz) consisting of four NMR lines with chemical shifts as indicated. The one at 11 ppm can probably be assigned to a small amount of LaAlO₃ which is not yet visible in the XRPD pattern while the other NMR lines reflect the different sites of the Al ions in the LLZ. An assignment of this NMR line to $\text{La}_2\text{Li}_{0.5}\text{Al}_{0.5}\text{O}_4$ is improbable due to the very low intensity of the ²⁷Al MAS NMR signal of this phase (see Fig. S1) combined with its small content as impurity phase in this sample.

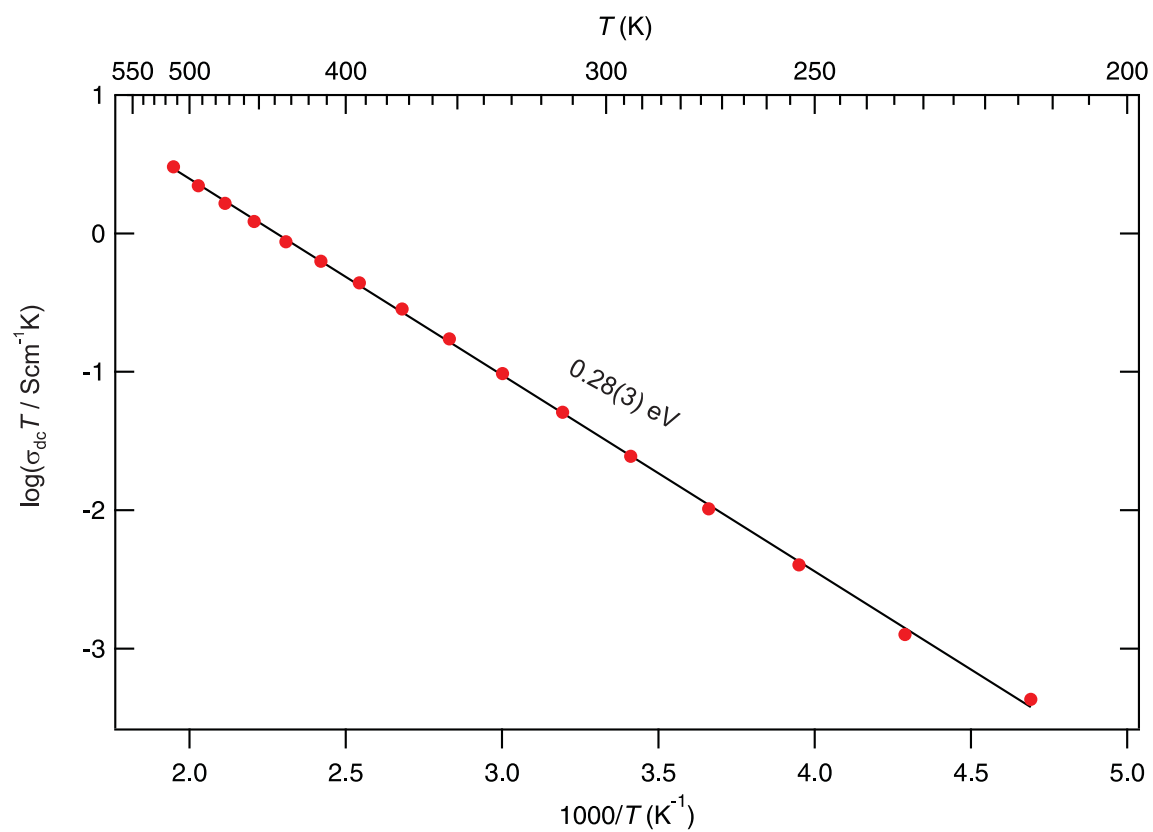


Fig. S3: Arrhenius plot of the dc conductivities of the sample of which the XRPD pattern and ^{27}Al MAS NMR spectrum are shown in Fig. S2. An activation energy of 0.28(3) eV was found.