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Mg²⁺ doping effect on ionic conductivity of Li_{1,15}Zr_{1,85}Y_{0,15}(PO₄)₃ Nasicon-type solid electrolyte for all-solid-state lithium ions batteries

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Solid electrolytes with Nasicon structure are already envisioned as promising Li⁺ conductive materials for all-solid-state batteries with good chemical stability in ambient atmosphere and wide electrochemical stability window [1] [2]. Li⁺ ionic conductivity of ~10⁻⁴ S/cm at room temperature was achieved for Li_{1,15}Zr_{1,85}Y_{0,15}(PO₄)₃ due to stabilization of better conductive rhombohedral phase (alpha) toward orthorhombic phase (beta) by Y³⁺ doping of Zr sites in the structure [2], [3]. Here we examine the impact of Mg doping and evaluate the potential Li⁺/Mg²⁺ conductivity in these structures for hybrid « Li⁺/Mg²⁺ » all-solid-state batteries, already demonstrated in conventional batteries with liquid electrolyte [4].

Several Mg²⁺ doped Nasicon samples were synthesized by modified Pechini route, and the gels were heated from 350°C to 1200°C under air with intermediate grindings. XRD was used to follow the synthesis and identify the formed phases depending on the doping level. ⁶Li and ³¹P MAS NMR will be used to evaluate the ions sites occupancy on the different samples. Ionic conductivity measurements were done by Electrochemical Impedance Spectroscopy on gold sputtered samples sintered by Spark Plasma Sintering method with high relative density (>95%).

We found that Mg²⁺ doping hinders the alpha phase formation, but at the same time both Li⁺ and Mg²⁺ are suspected to contribute to the conductivity in this solid electrolyte material. Further analysis will be held to study the Li⁺ and Mg²⁺ diffusions in these materials using post-mortem ion beam analysis on half-cells with Mg metal or Li metal anodes.

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