

Supporting Information

Low-temperature densification of Al-doped $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$: A reliable and controllable synthesis of fast-ion conducting garnets

Hany El-Shinawi, Gary W. Paterson, Donald MacLaren, Edmund J. Cussen and Serena A. Corr

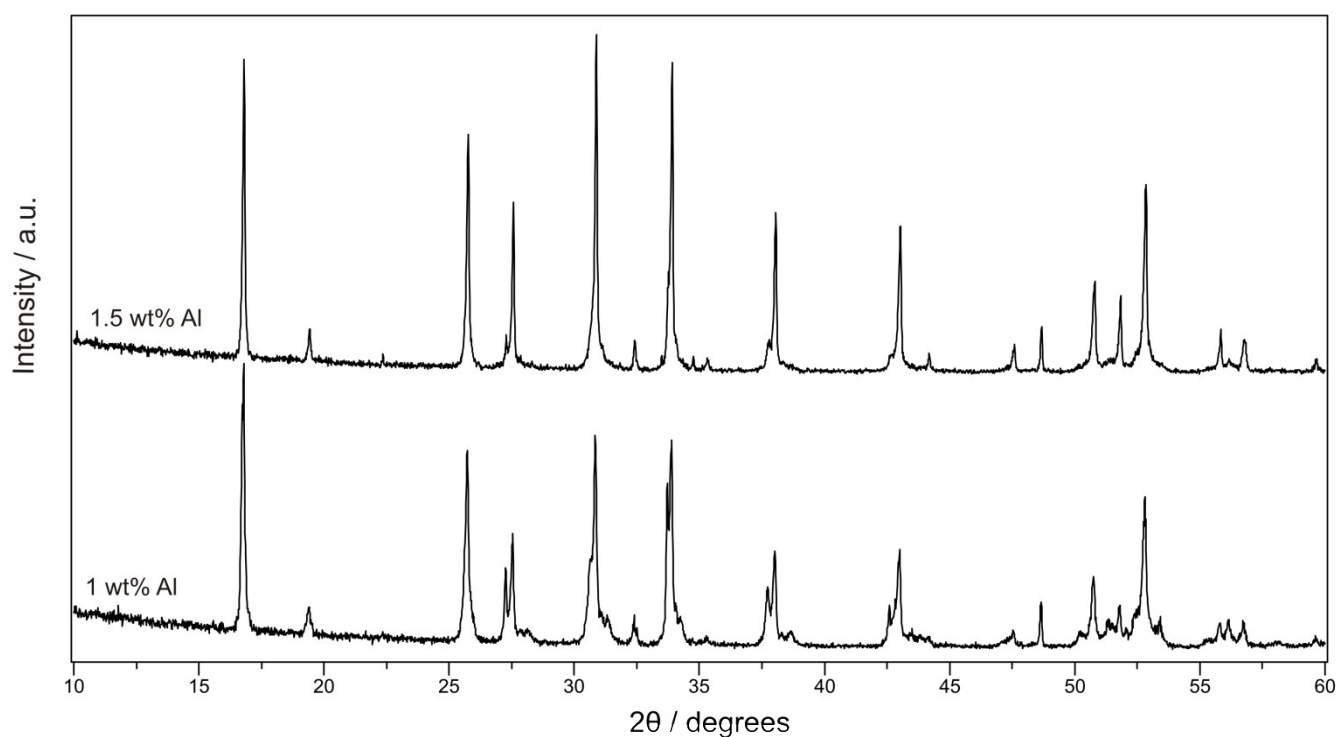


Figure S1. XRD patterns for LLZO phases containing 1.0 and 1.5 wt% Al. These phases contain stoichiometric amounts of Zr (*i.e.* no excess of Zr). The impurity phases LiAlO_2 and Li_2ZrO_3 are not seen, but LLZO exhibits peak broadening/splitting.

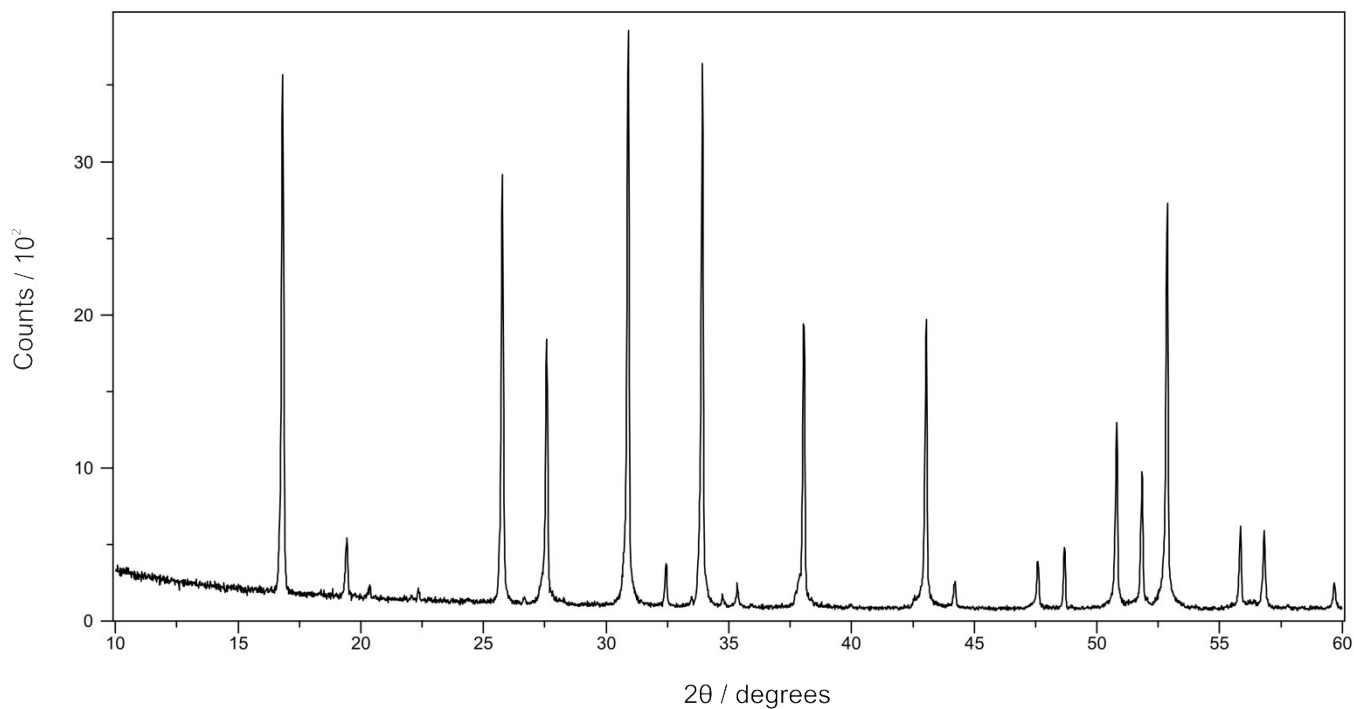


Figure S2. XRD pattern for LLZO containing excess of Zr and doped with 1.5 wt % Al. The impurity phases LiAlO_2 and Li_2ZrO_3 are seen, and the LLZO diffraction peaks exhibit some broadening.

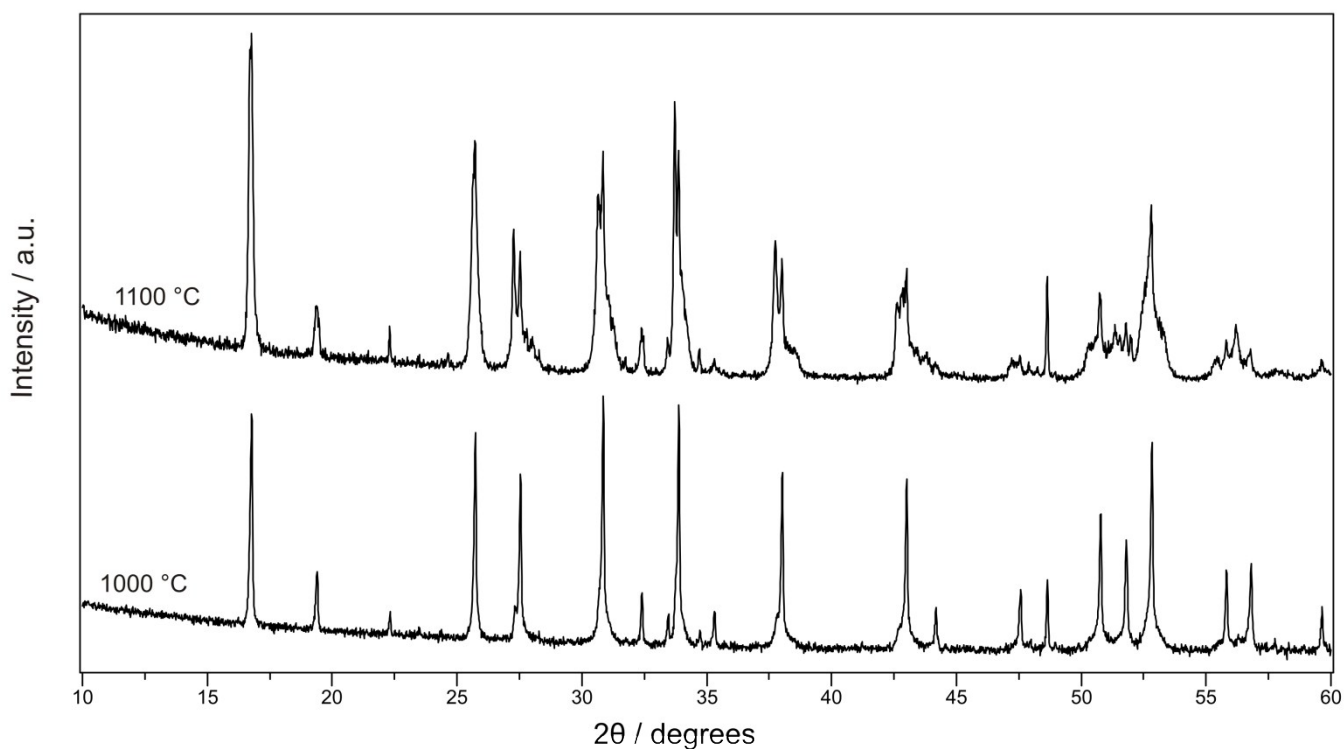


Figure S3. XRD patterns for LLZO phases containing stoichiometric amounts of Zr (*i.e.* no excess of Zr) and doped with 2 wt% Al. The impurity phase Li_2ZrO_3 is not observed, but LLZO exhibits peak broadening/splitting, particularly at higher calcination temperature.

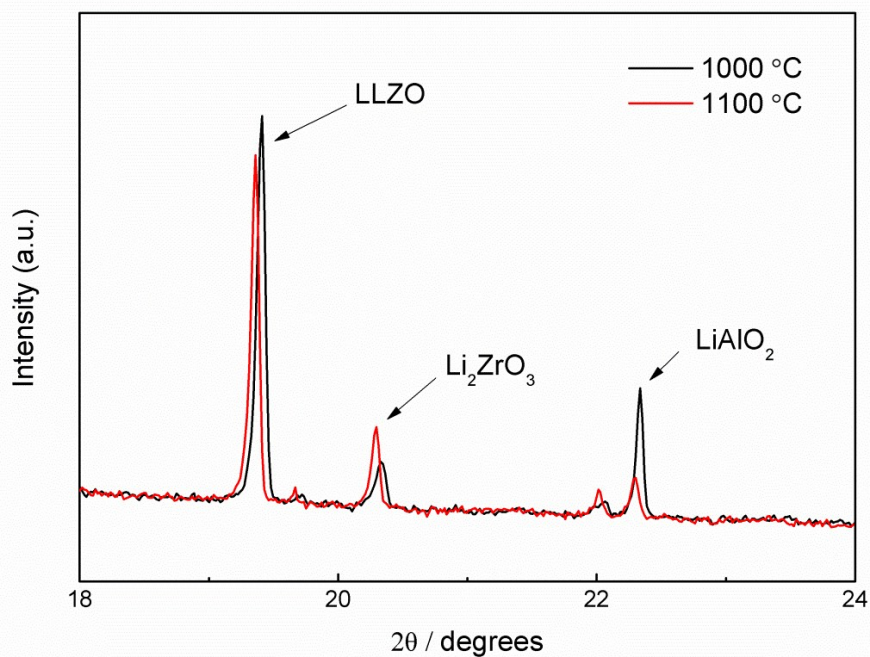


Figure S4. XRD patterns (2θ from 18 to 24°) of LLZO (2 wt% Al) calcined at 1000 and 1100 °C.

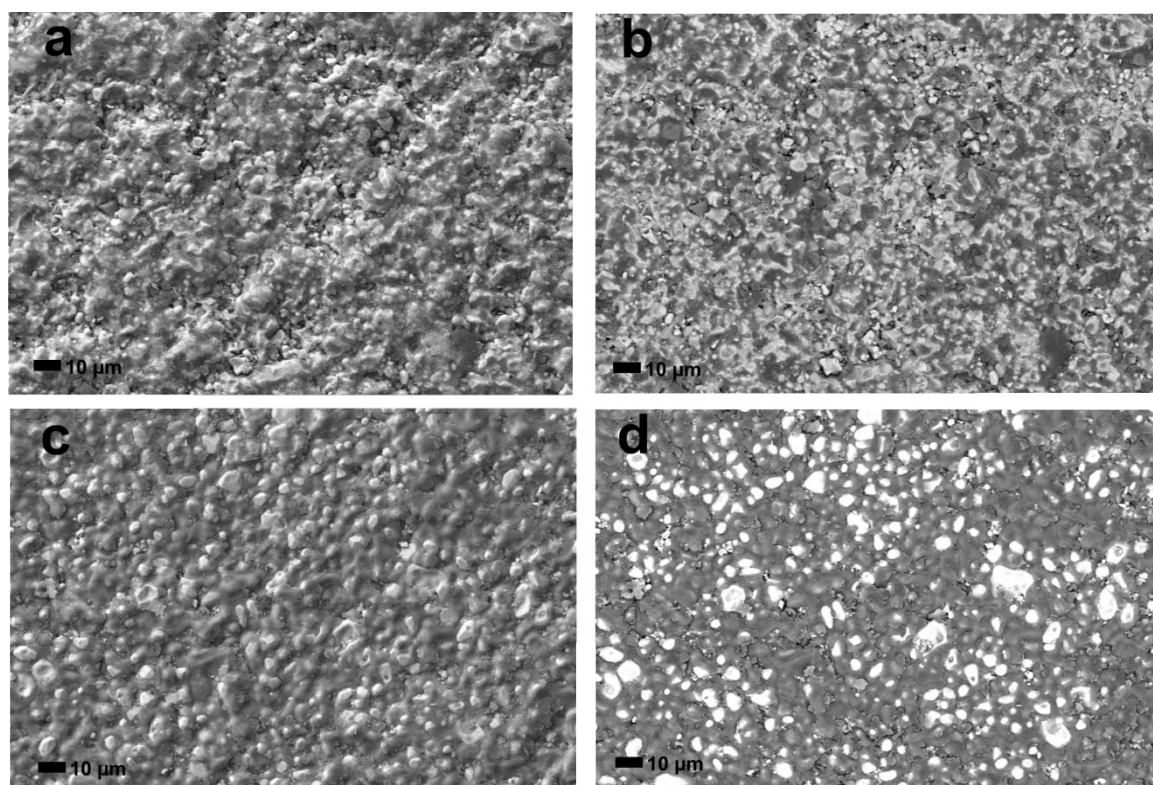


Figure S5. SEM images of the surface of the pellets calcined at 1000 °C (a,b), and 1100 °C (c,d). The images b and d were collected using an AsB detector.

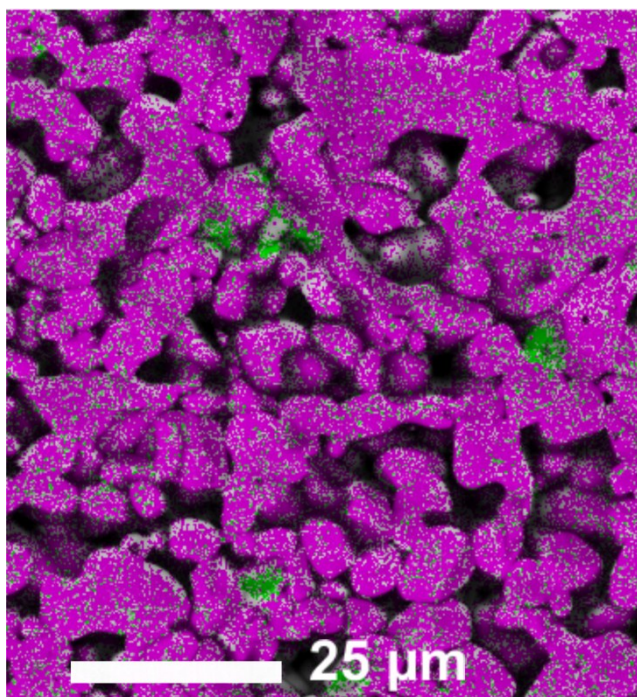


Figure S6. EDS elemental map of the bulk of the pellet showing La (red) and Zr (green) distribution. The La-free particles (fully green) are Li_2ZrO_3 particles.

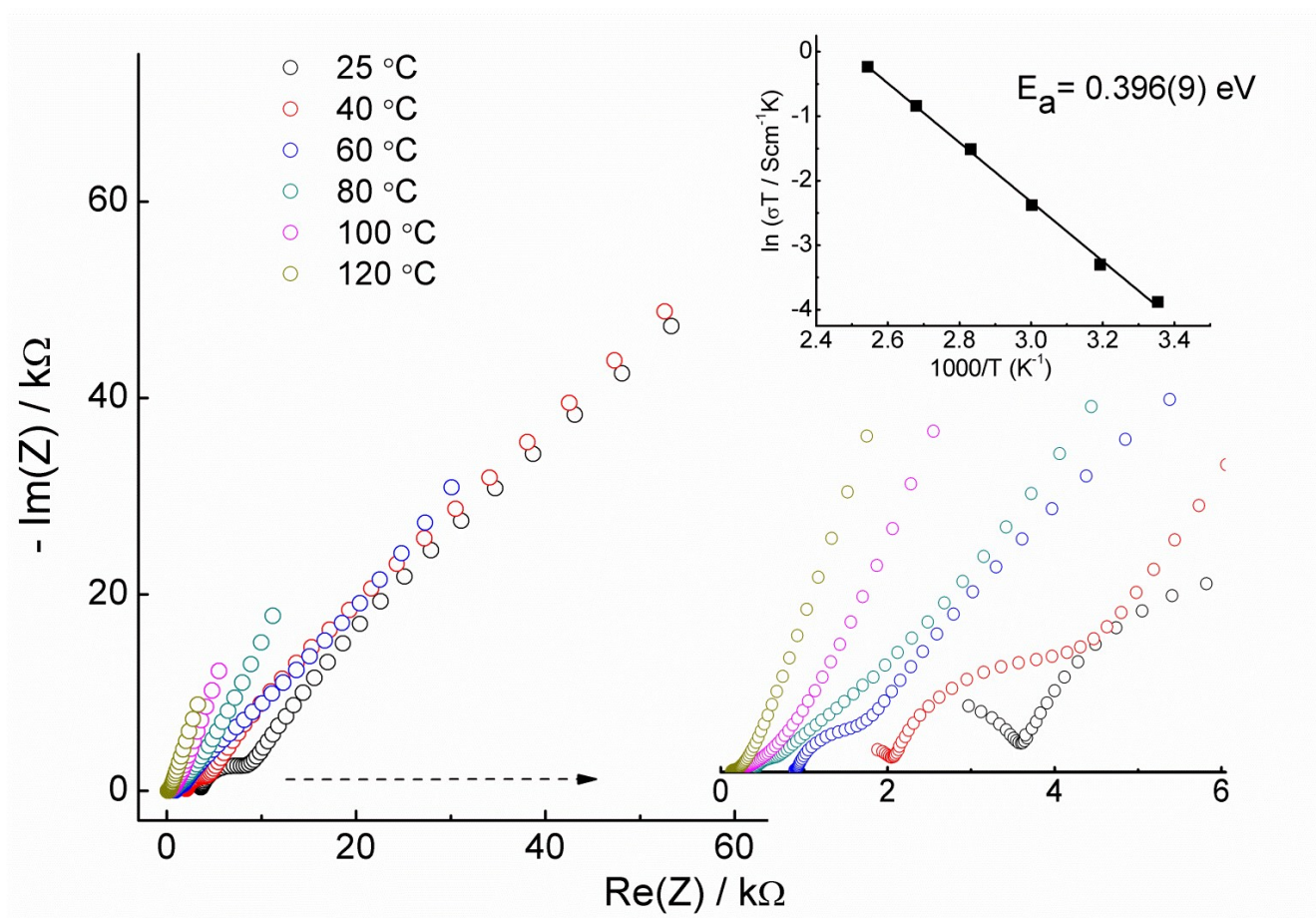


Figure S7. Impedance spectra and Arrhenius plot for the total conductivity of LLZO (2 wt% Al) calcined at 1000 °C in the temperature range 25-120 °C.

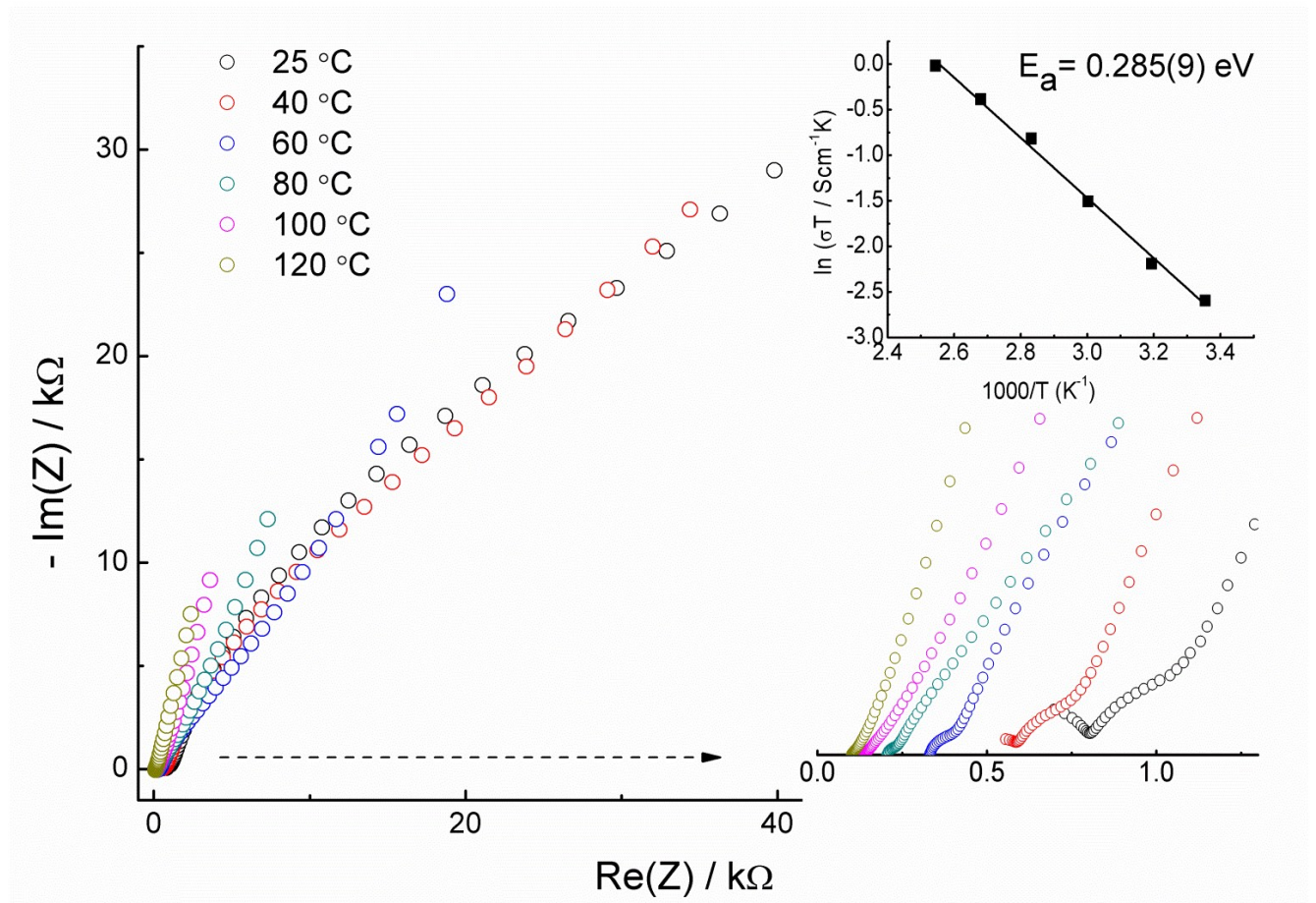


Figure S8. Impedance spectra and Arrhenius plot for the total conductivity of LLZO (2 wt% Al) calcined at 1100 °C (polished pellet) in the temperature range 25-120 °C.

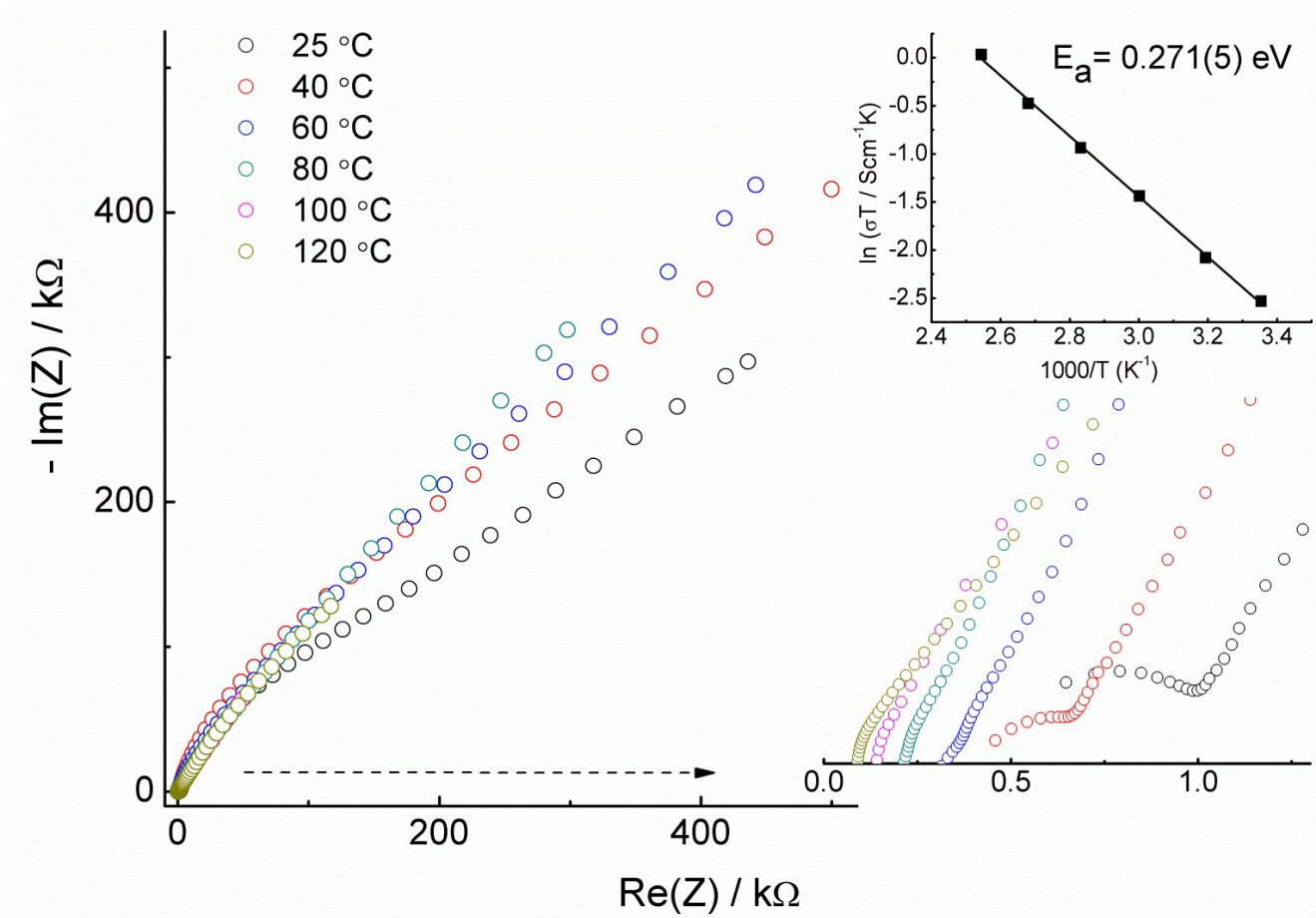


Figure S9. Impedance spectra and Arrhenius plot for the total conductivity of LLZO (2 wt% Al) calcined at 1100 °C (unpolished pellet) in the temperature range 25-120 °C.

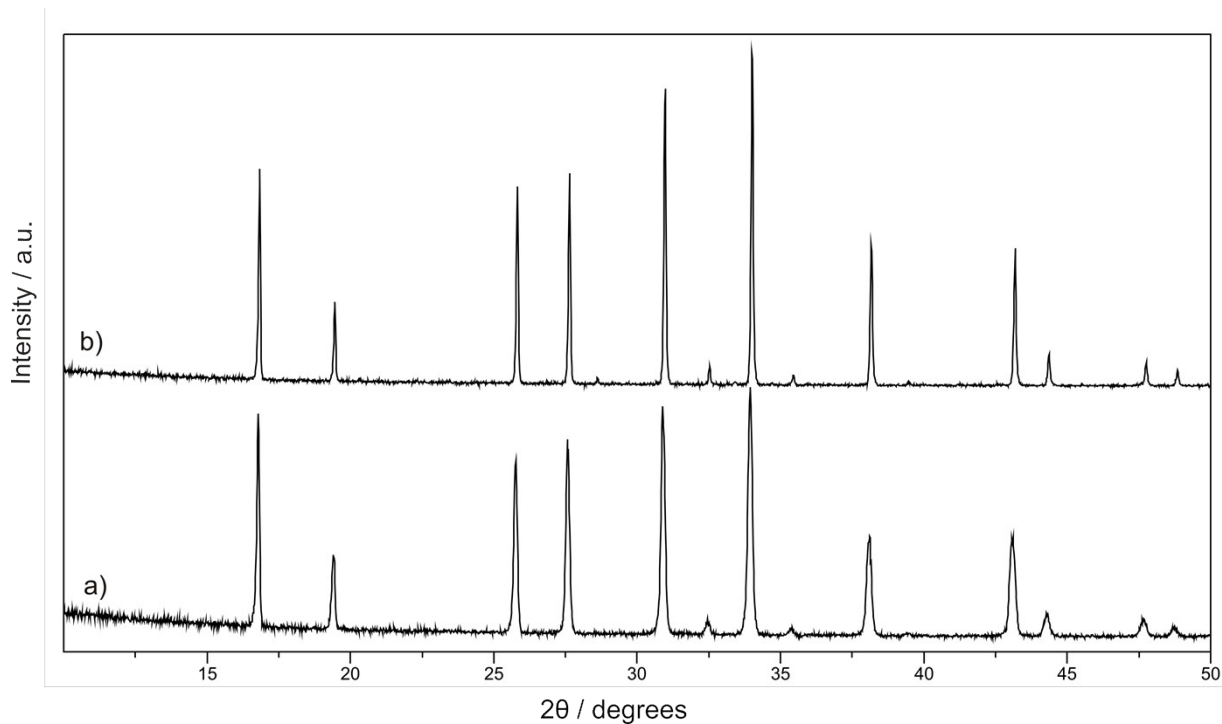


Figure S10. a) XRD pattern of $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ prepared by sol-gel method and calcined at $1000\text{ }^\circ\text{C}$ for 2h. b) XRD pattern of $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ mixed with 1 wt% Al and calcined at $1000\text{ }^\circ\text{C}$ for 2h.

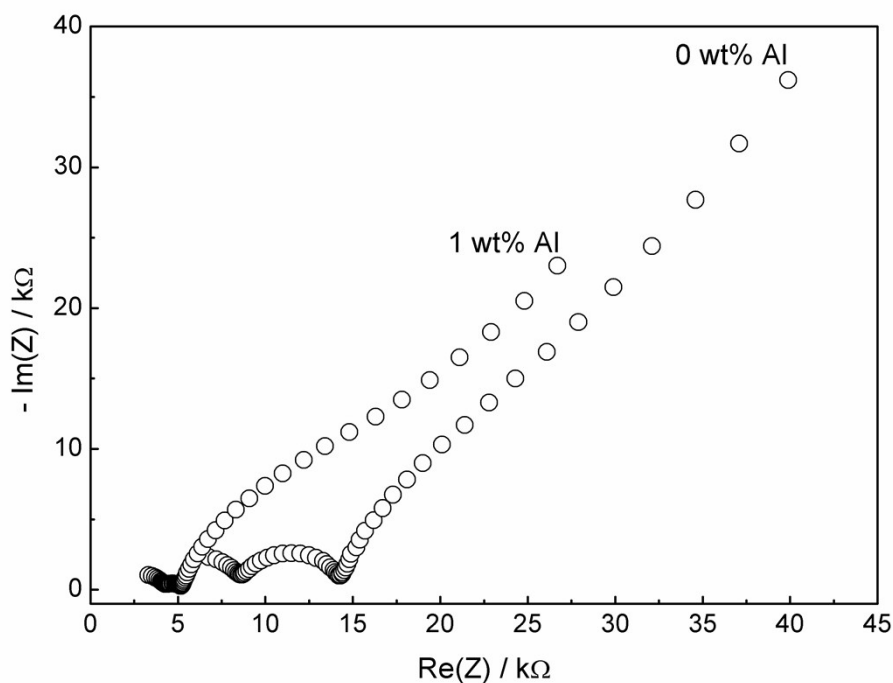


Figure S11. Impedance spectra for $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ densified at $1000\text{ }^\circ\text{C}$ in absence and in the presence of Al additives. The spectra were collected at room temperature using gold electrodes ($1\text{-}10^6$ Hz frequency range), and show the improvement of the ionic conductivity after mixing with Al_2O_3 nanosheets.