

## Supporting Information for Journal of Materials Chemistry A

### Study of Thermal Material Parameters for Ta and Al Substituted $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) Solid State Electrolyte in Dependency of Temperature and Grain Size

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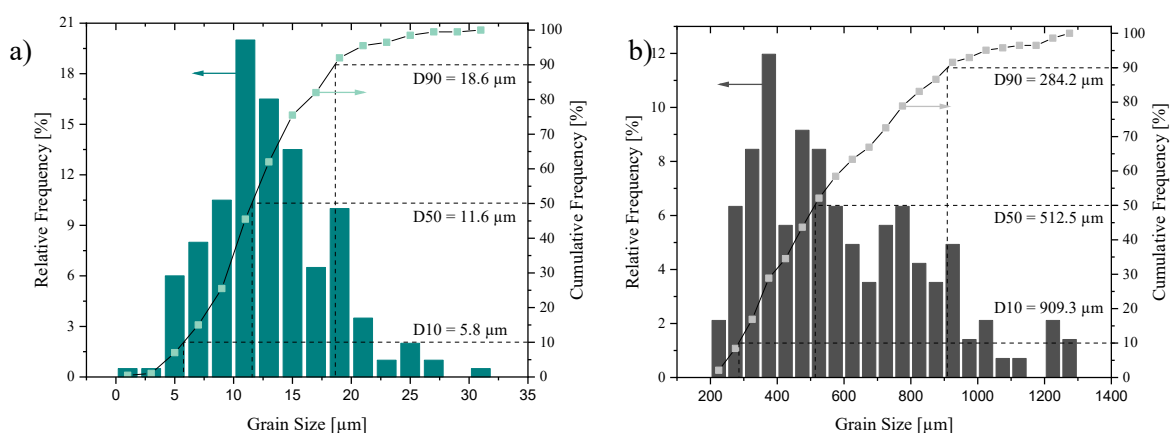


Fig. S1: Relative and cumulative grain size distribution with indicated D10, D50 and D90 percentile values of a) LLZO:Ta and b) LLZO:Al.

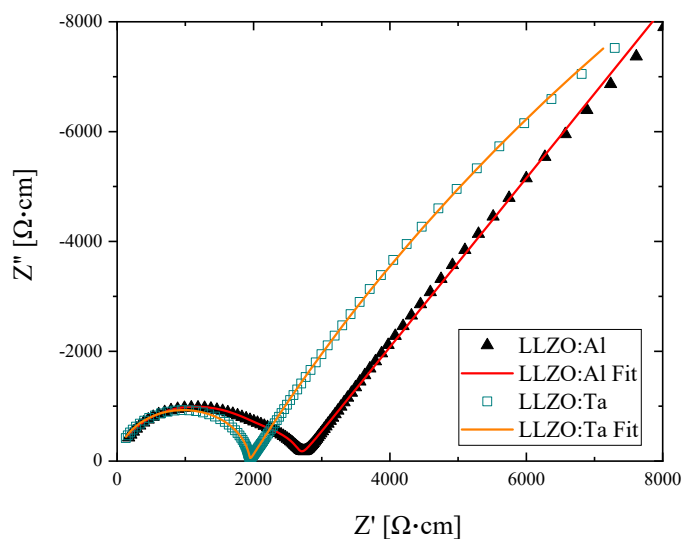
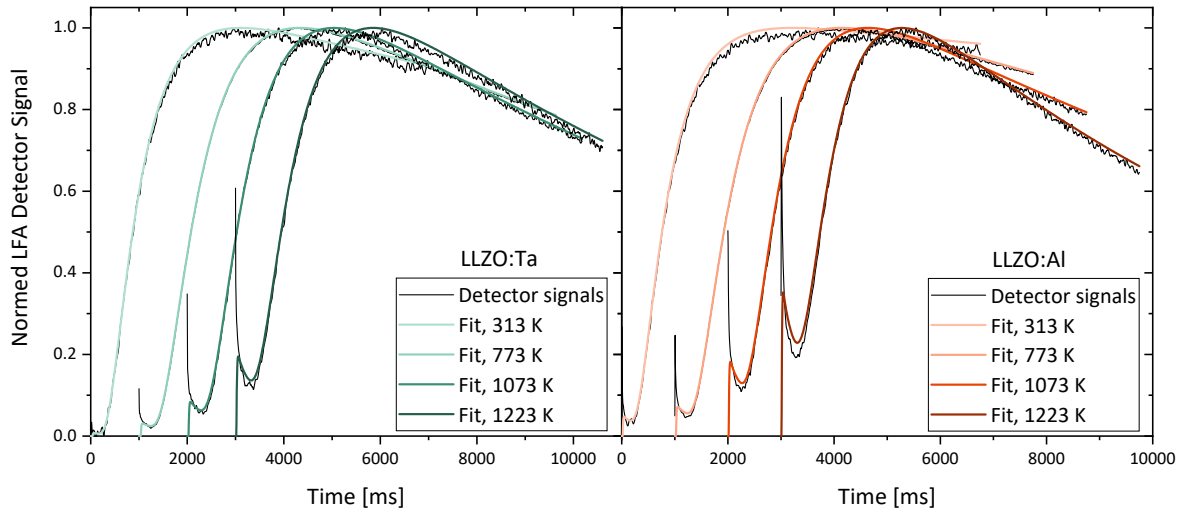
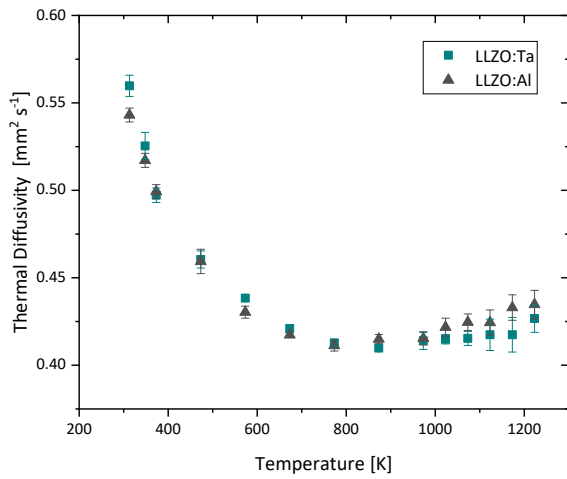


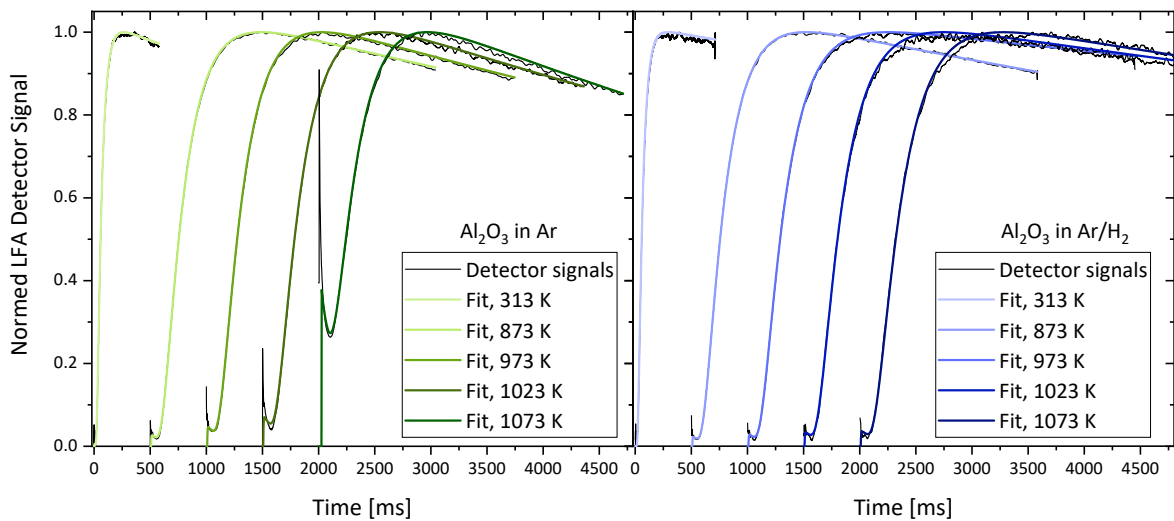
Fig. S2: Nyquist plot for LLZO:Ta and LLZO:Al. Data are normalized to the ratio of sample thickness to sample area.



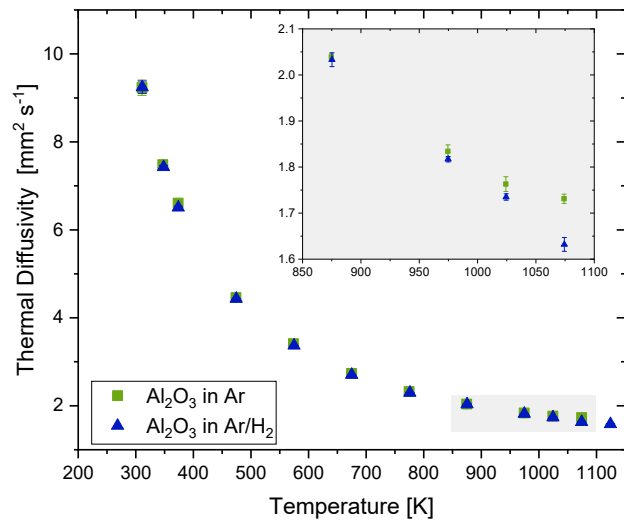
**Fig. S3:** Normed laser flash detector signals with corresponding fits (transparent (radiation) model<sup>35</sup>) for LLZO:Ta (left) and LLZO:Al (right) at 313 K, 773 K, 1073 K, and 1223 K. The signals at 773 K, 1073 K, and 1223 K are each shown with a time offset of 1000 ms for better clarity.



**Fig. S4:** Temperature-dependent thermal diffusivity of LLZO:Ta and LLZO:Al (both porosity corrected) determined by laser flash analysis under Ar/H<sub>2</sub> atmosphere. The shown data are the mean of five measurements with standard deviation shown as error bars.



**Fig. S5:** Normed laser flash detector signals with corresponding fits (transparent (radiation) model<sup>35</sup>) for an Al<sub>2</sub>O<sub>3</sub> reference sample under Ar-atmosphere (left) and Ar/H<sub>2</sub>-atmosphere (right) at 313 K, 873 K, 973 K, 1023 K, and 1073 K. The signals at 873 K, 973 K, 1023 K, and 1073 K are each shown with a time offset of 500 ms for better clarity.



**Fig. S6:** Temperature-dependent thermal diffusivity of an Al<sub>2</sub>O<sub>3</sub> reference sample determined by laser flash analysis under Ar and Ar/H<sub>2</sub> atmosphere. The shown data are the mean of three measurements with standard deviation shown as error bars. Inset: Closeup to the temperature range between 850 and 1100 K.