Unraveling the Role of Aluminum in Boosting Lithium-Ionic Conductivity of LLZO

Table S1. Ionic conductivity and activation energy of pure LLZO and doped LLZO.

LLZO type	Ionic Conductivity, S/cm	Activation Energy, eV
c-LLZO ($Li_7La_3Zr_2O_{12}$)	2.38E-03	0.227109818
t-LLZO ($Li_7La_3Zr_2O_{12}$)	3.03E-07	0.495565601
$Li_{44}Al_4La_{24}Zr_{16}O_{96}$	1.44E-02	0.13823926
$Li_{44}Ga_4La_{24}Zr_{16}O_{96}$	3.73E-03	0.196992238
$Li_{58}La_{23}RbZr_{16}O_{96}$	3.32E-03	0.211814052
$Li_{48}La_{24}Zr_8Ta_8O_{96}$	7.12E-03	0.191399589
$Li_{50}Fe_2La_{24}Zr_{16}O_{96}$	5.80E-03	0.205600954
$Li_{53}In_1La_{24}Zr_{16}O_{96}$	3.25E-03	0.227368338
$Li_{53}As_1La_{24}Zr_{15}O_{96}$	2.63E-03	0.201257818
$Li_{54}La_{22}Ge_2Zr_{16}O_{96}$	2.58E-03	0.218492485

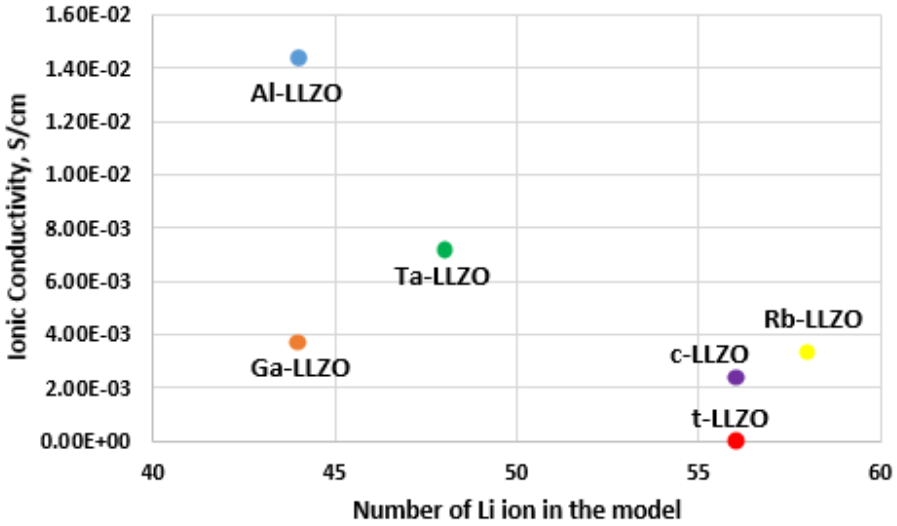


Figure S1. Ionic conductivity with corresponding number of Li present in the model.

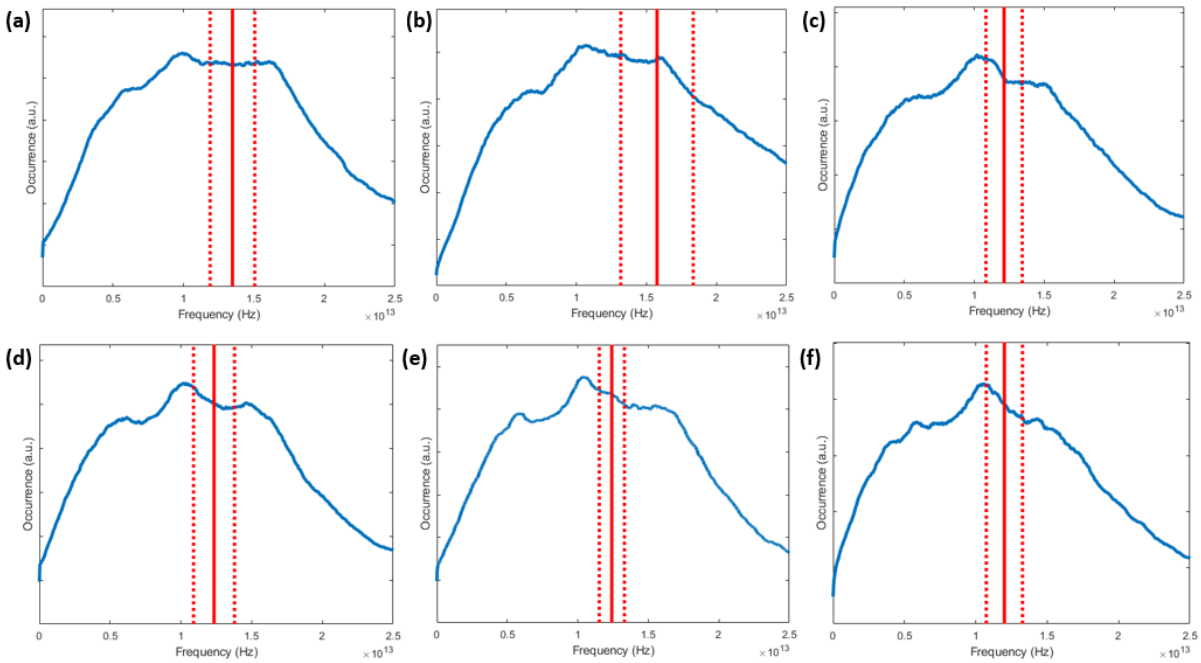


Figure S2. Spectrum of vibrational frequency of (a) c-LLZO (b) t-LLZO (c) Al-LLZO (d) Ga-LLZO (e) Rb-LLZO (f) Ta-LLZO at 1000 K, the attempt frequency is shown in solid red line and the red dotted lines are indicating standard deviation.

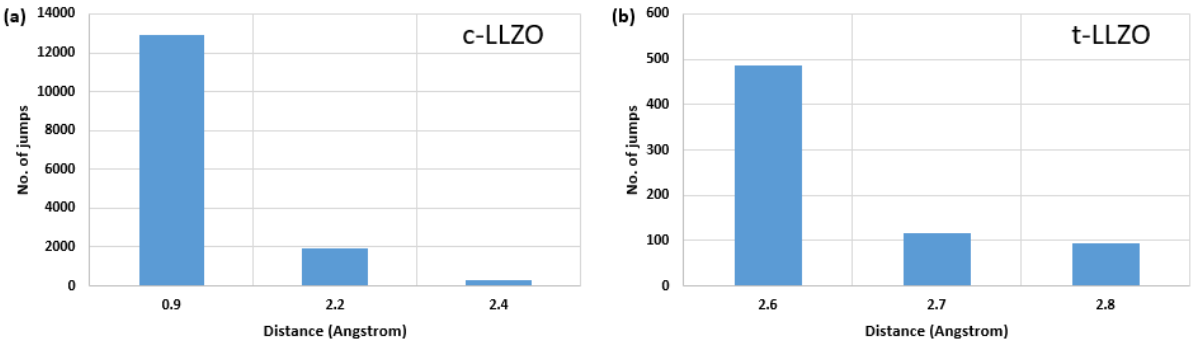


Figure S3. Number of successful jumps in (a) c-LLZO and (b) t-LLZO during MD simulation at 1500 K.