

Supplementary Materials

Towards improved solid state Li electrolyte: A first principles-based investigation on structure, migration pathways of Li ions and Li ionic conductivity of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$

Muhammad Mozammel Kamal Raju ¹, Yulun Han ², Dmitri Kilin ^{2,3,*}, Yi Ding ^{4,*}, and Qifeng Zhang ^{1,3,*}

¹Department of Electrical and Computer Engineering, North Dakota State University, Fargo, ND 58108, USA

²Department of Chemistry and Biochemistry, North Dakota State University, Fargo, ND 58108, USA

³The Materials and Nanotechnology (MNT) Program, North Dakota State University, Fargo, ND 58105, USA

⁴Ground Vehicle Systems Center, Located (GVSC), US Army Combat Capabilities Development Command (DEVCOM), Warren, MI 48397-5000, USA

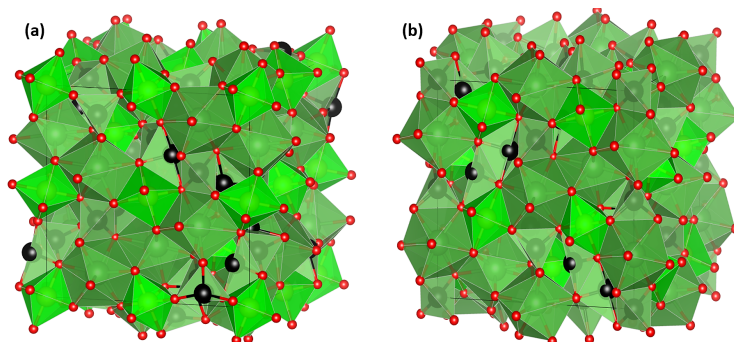


Figure S1. Polyhedral view of (a) cubic, and (b) tetragonal LLZO ($\text{Li}_{56}\text{La}_{24}\text{Zr}_{16}\text{O}_{96}$); red and black spheres represent oxygen (O) and Lithium (Li) respectively; in which La and Zr ions are located in the center of dark-green dodecahedrons and light-green octahedrons, respectively where O atoms are forming polyhedrons with La and Zr ions.

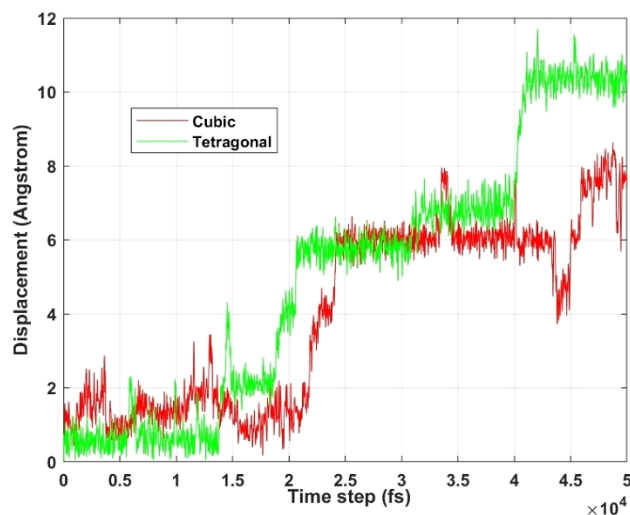
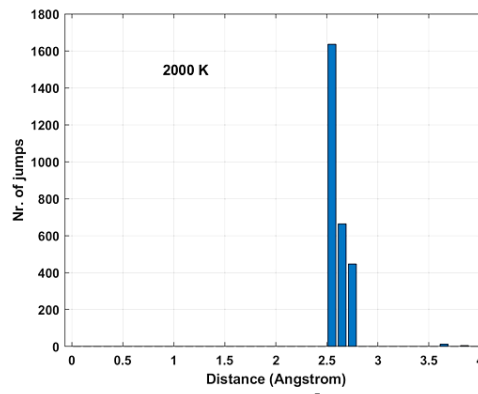
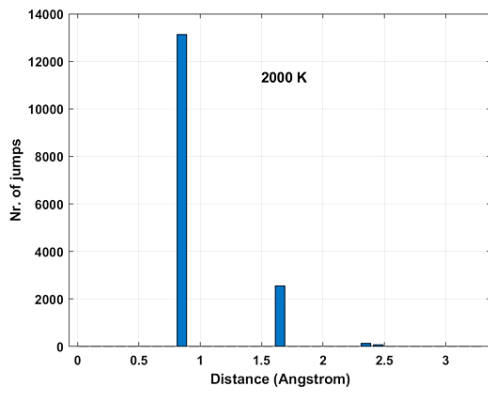
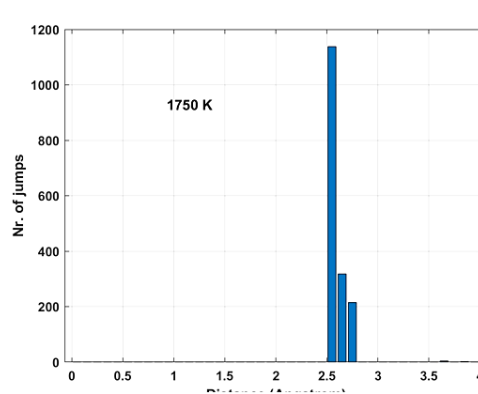
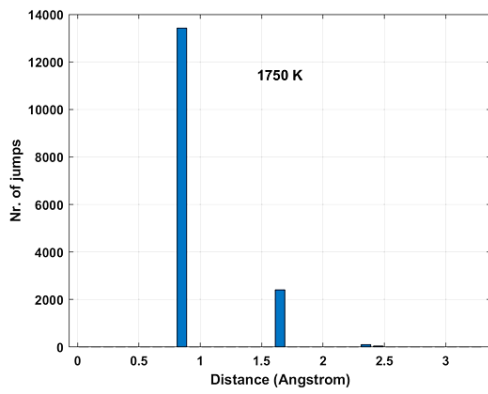
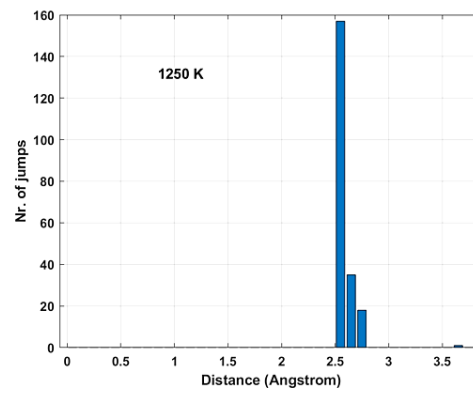
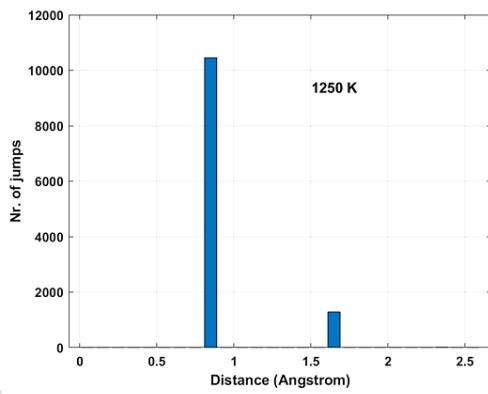
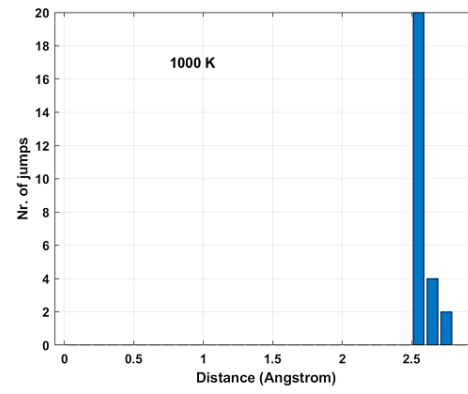
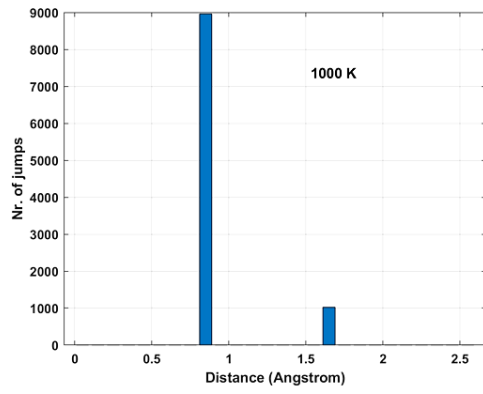


Figure S2. Total displacement covered by Li(54) ion at 1500 K.



Cubic LLZO

Tetragonal LLZO

Figure S3. Number of successful jumps in cubic and tetragonal LLZO during MD simulation at different temperatures.

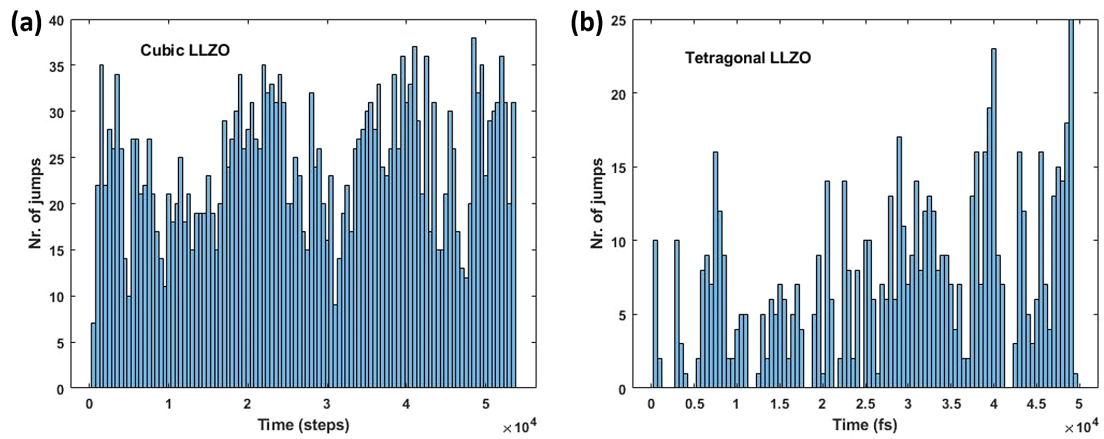


Figure S4. Li ions jump histograms during the MD simulation at 1500 K.

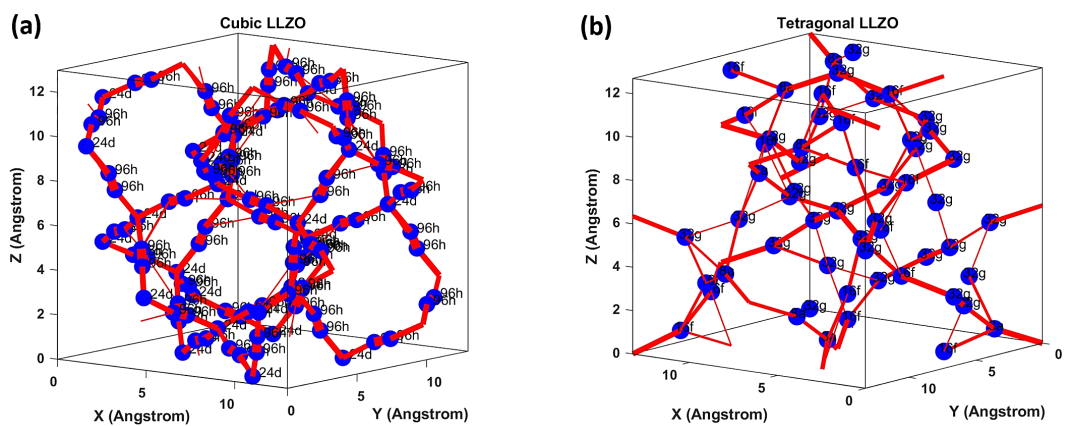


Figure S5. Li ions transition between different sites during the MD simulation at 1500 K.

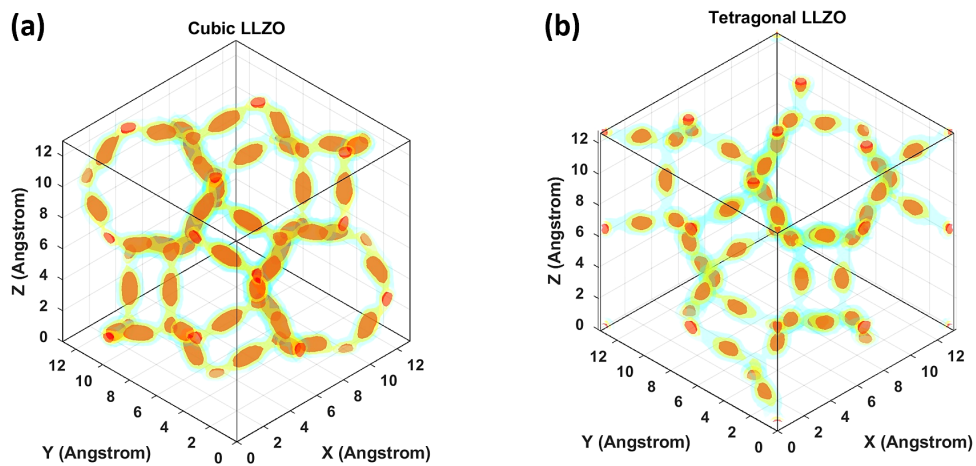


Figure S6. Density of diffusion and loop structured pathway followed by Li ions during the MD simulation at 1500 K.

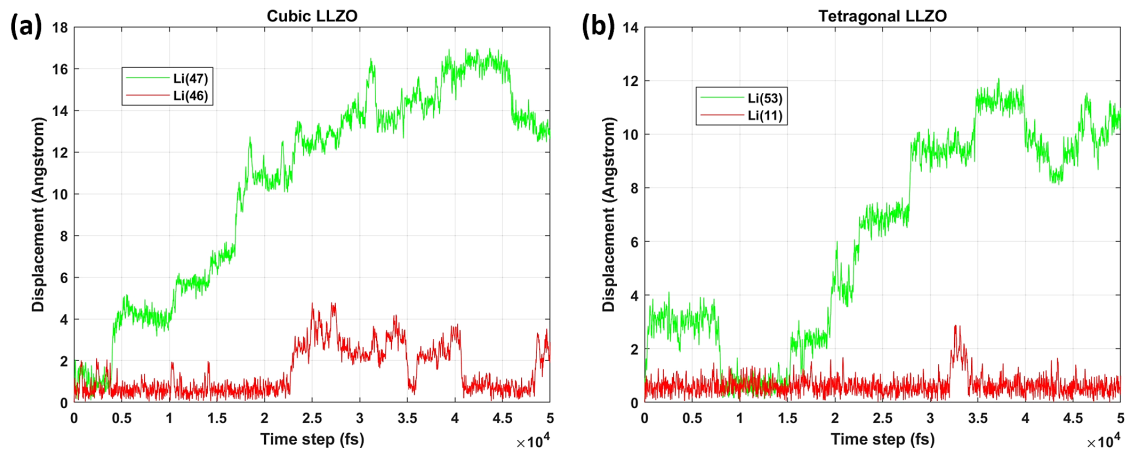


Figure S7. Shortest and longest distance covered by Li ions during the MD simulation at 1500 K.

Table S1. Calculated diffusion coefficients and logarithms of diffusion coefficient at different temperatures.

Temperature	Phase	750 K	1000 K	1250 K	1500 K	1750 K	2000 K
Diffusion coefficient, D (m ² /s)	Cubic	3.33×10^{-10}	8.33×10^{-10}	8.33×10^{-10}	1.67×10^{-09}	2.67×10^{-09}	3.17×10^{-09}
	Tetragonal	3.33×10^{-11}	5.00×10^{-11}	5.00×10^{-10}	1.00×10^{-09}	2.00×10^{-09}	2.50×10^{-09}
ln(D) (m ² /s)	Cubic	-21.8219	-20.9056	-20.9056	-20.2124	-19.7424	-19.5706
	Tetragonal	-24.1245	-23.719	-21.4164	-20.7233	-20.0301	-19.807