Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting Information

Non-equilibrium Kinetics for improving Ionic Conductivity in Garnet Solid Electrolyte

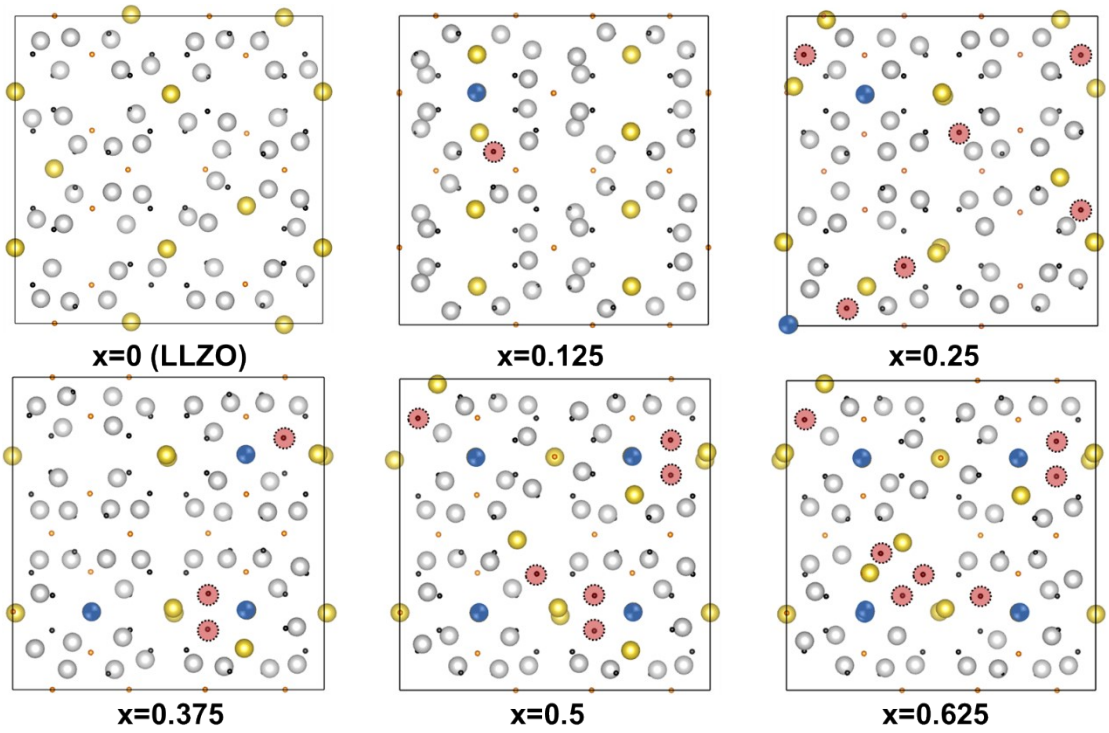

Figure S1 Li_{24d} (yellow), Li_{48g} (gray), V_{48g} (red) and Ta (dark blue) ions distribution in $Li_{7-x}La_3Zr_{2-x}Ta_xO_{12}$ (x=0, 0.125, 0.25, 0.375, 0.5 and 0.625) crystal.

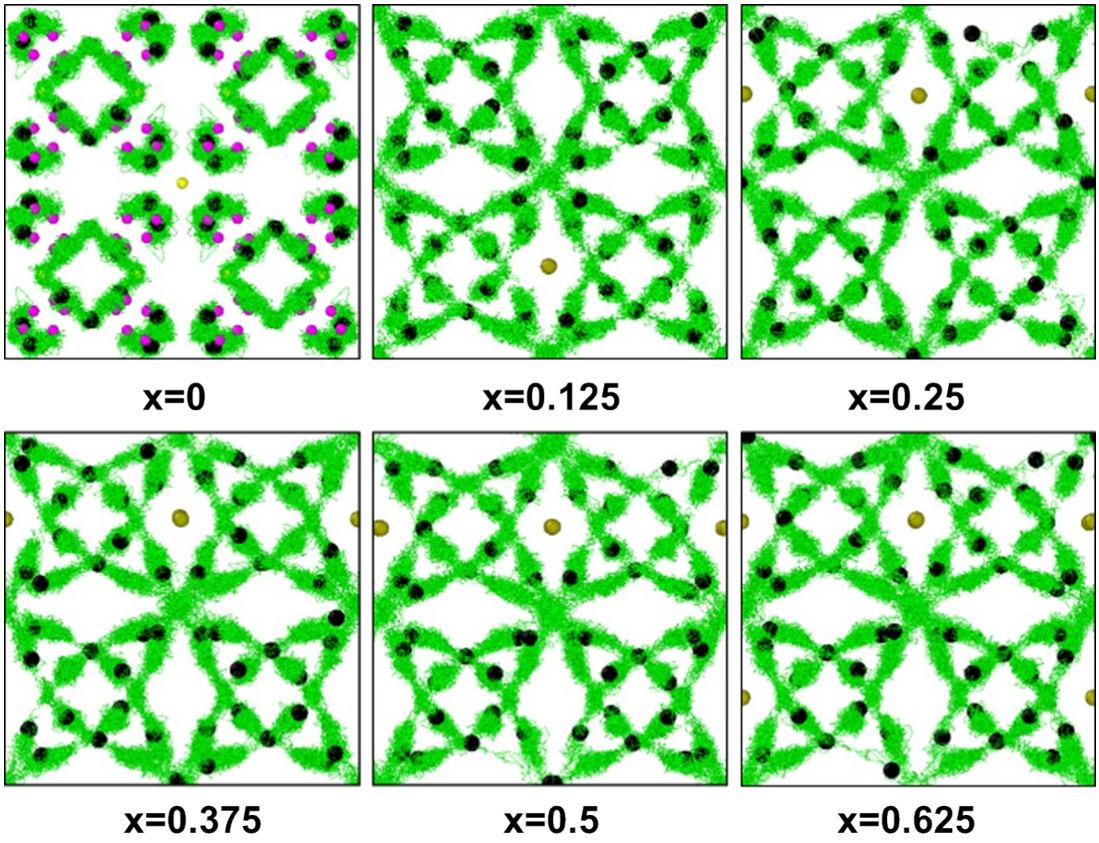

Figure S2 Lithium trajectories with continual Ta adoptions in $Li_{7-x}La_3Zr_{2-x}Ta_xO_{12}$ (x=0, 0.125, 0.25,0.375, 0.5 and 0.625) by AIMD calculations.

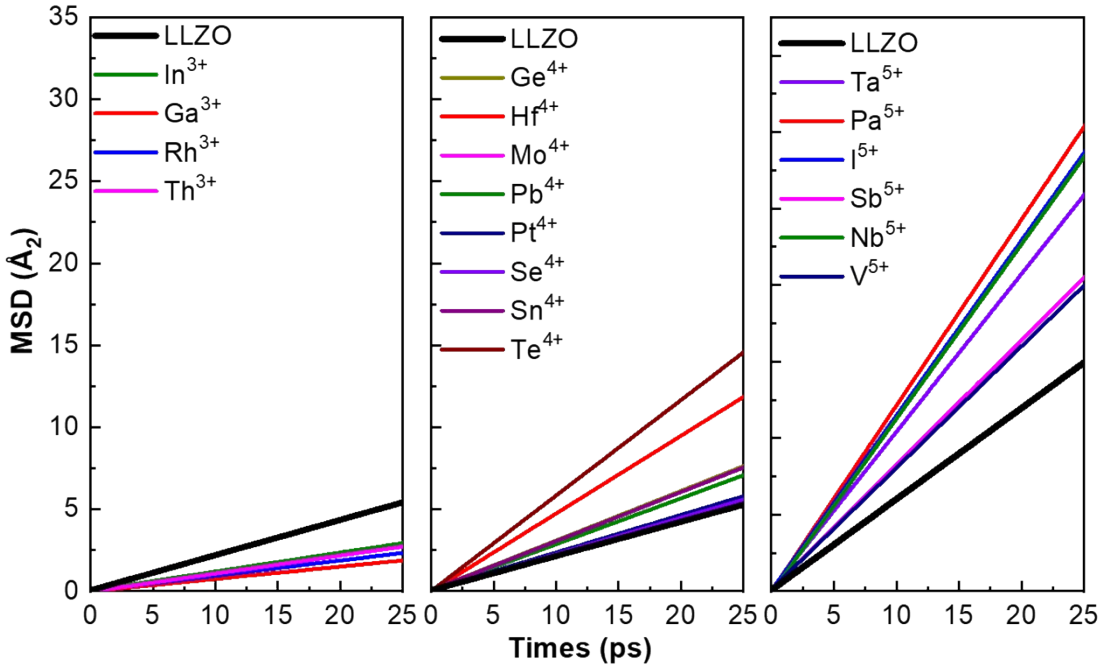

Figure S3 MSD plots of Li^{+} in doped $Li_{7\pm y}La_3Zr_{1.875}M_{0.125}O_{12}$ at 800 K in AIMD simulations. M is tri-valent, quadri-valent or pentad-valent element. The subscript of y varies to maintain electro-neutrality.

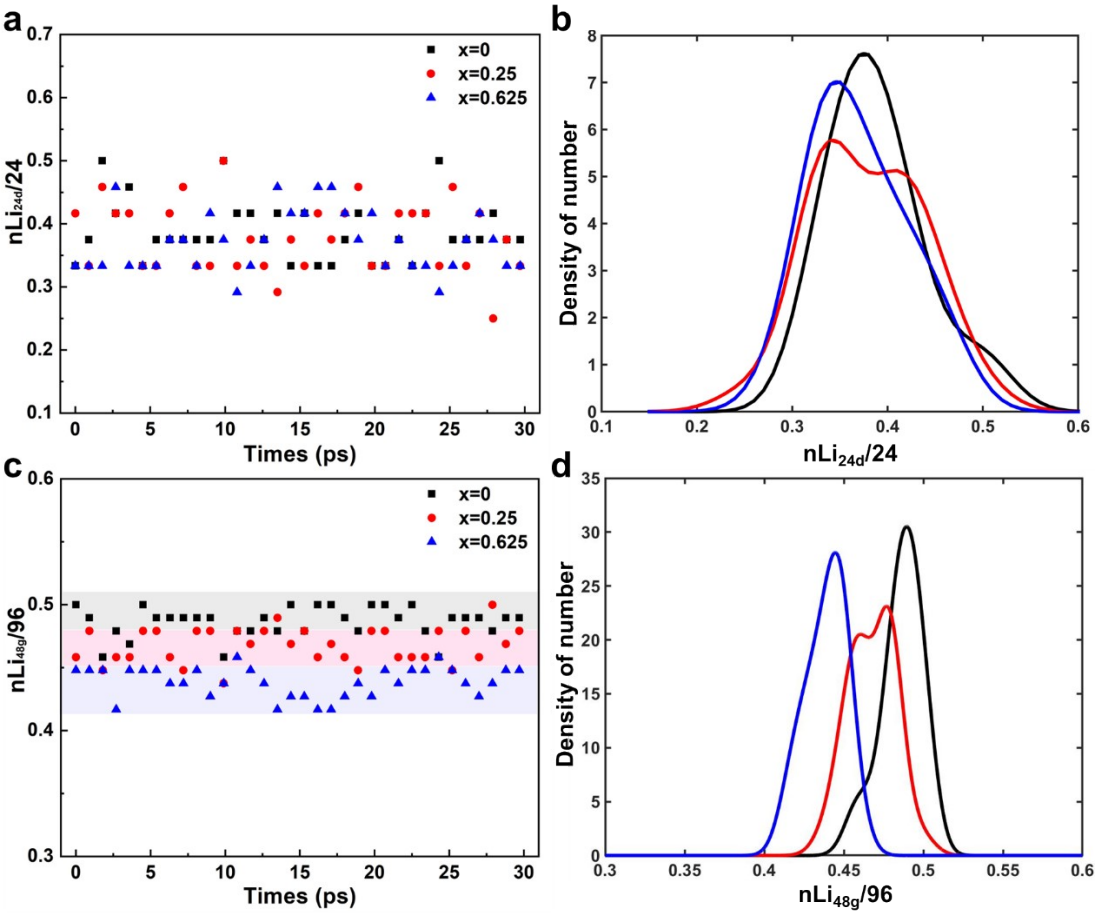

Figure S4 (**a**) Occupancy ratio and (**b**) density of distribution of Li_{24d} in $Li_{7-x}La_3Zr_{2-x}Ta_xO_{12}$ (x=0, 0.25, 0.625) per ps during MD running at 1200 K for 30ps. (**c**) Occupancy ratio and (**d**) density of distribution of Li_{24d} in $Li_{7-x}La_3Zr_{2-x}Ta_xO_{12}$ (x=0, 0.25, 0.625) per ps during MD running at 1200 K for 30ps.

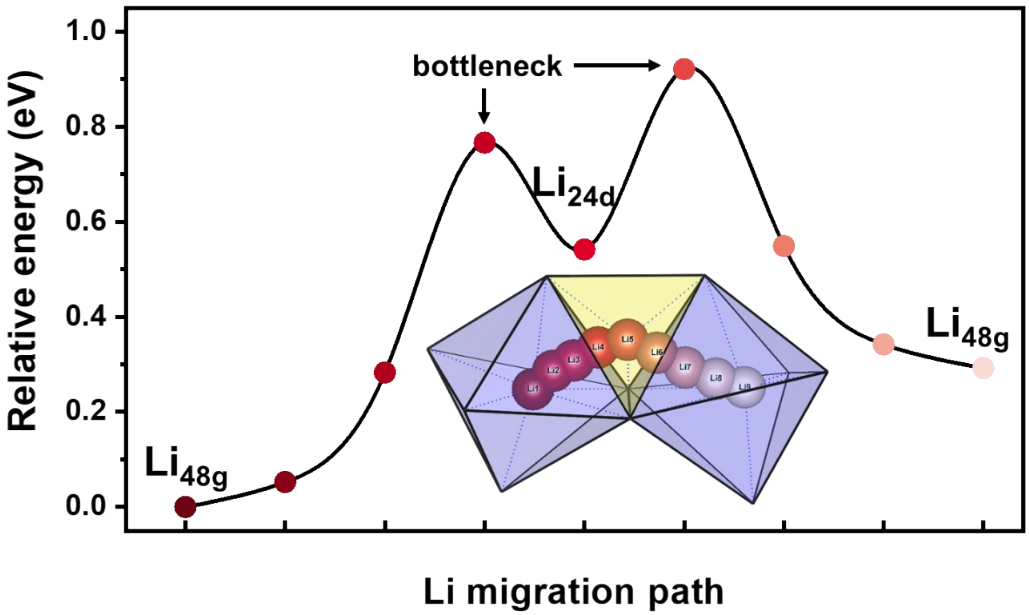

Figure S5 Single-ion migration of Li^+ following the Li_{48g}→Li_{24d}→Li_{48g} trajectory.