

Supporting Information

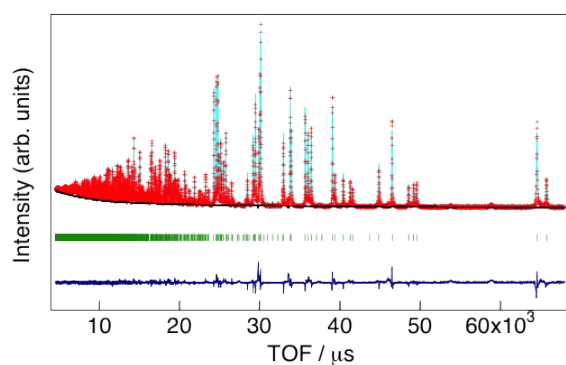


Fig. S1. Observed, calculated and difference plots for the Rietveld analysis of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$.

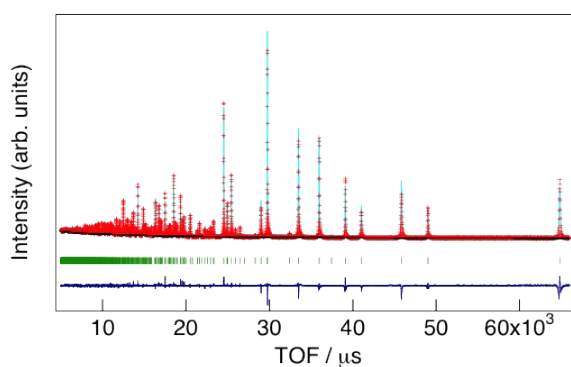


Fig. S2. Observed, calculated and difference plots for the Rietveld analysis of $\text{Li}_{6.6}\text{La}_3\text{Zr}_{1.6}\text{Ta}_{0.4}\text{O}_{12}$.

Table S1. Atomic coordinates of Li₇La₃Zr₂O₁₂.

Atom	Site	<i>g</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> / Å ²
Li1	8 <i>a</i>	1	0	1/4	3/8	1.04(4)
Li2	16 <i>f</i>	1	0.17949(9)	= <i>x</i> (Li2)+1/4	1/8	1.44(3)
Li3	32 <i>g</i>	1	0.08113(9)	0.08748(9)	0.80474(9)	1.10(2)
La1	8 <i>b</i>	1	0	1/4	1/8	0.371(7)
La2	16 <i>e</i>	1	0.12722(2)	0	1/4	0.385(5)
Zr	16 <i>c</i>	1	0	0	0	0.334(5)
O1	32 <i>g</i>	1	-0.03440(3)	0.05513(2)	0.15273(3)	0.516(5)
O2	32 <i>g</i>	1	0.05416(3)	0.85237(3)	0.53382(3)	0.539(6)
O3	32 <i>g</i>	1	0.14992(3)	0.02765(3)	0.44702(3)	0.536(6)

Space group: *I4*₁/*acd* (142-2): *a* = 13.1247(11) Å, *c* = 12.6640(16) Å, *R*_{wp} = 6.69%, *R*_p = 5.28%, *R*_B = 4.16%, *R*_F = 4.00%, χ^2 = 5.16.

Table S2. Atomic coordinates of Li_{6.6}La₃Zr_{1.6}Ta_{0.4}O₁₂.

Atom	Site	<i>g</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> / Å ²
Li1	24 <i>d</i>	0.367(6)	3/8	0	1/4	2.11(8)
Li2	96 <i>h</i>	0.456(3)	0.0973(16)	0.6886(18)	0.5789(16)	1.48(6)
La1	24 <i>c</i>	1	1/8	0	1/4	0.436(7)
Zr	16 <i>a</i>	0.8	0	0	0	= <i>B</i> (Ta)
Ta	16 <i>a</i>	0.2	0	0	0	0.436(7)
O1	96 <i>h</i>	1	0.28148(2)	0.10117(2)	0.19656(2)	0.857(7)

Space group: *Ia*-3*d* (230): *a* = 12.94191(10) Å, *R*_{wp} = 8.91%, *R*_p = 4.56%, *R*_B = 3.34%, *R*_F = 3.26%, χ^2 = 3.81.