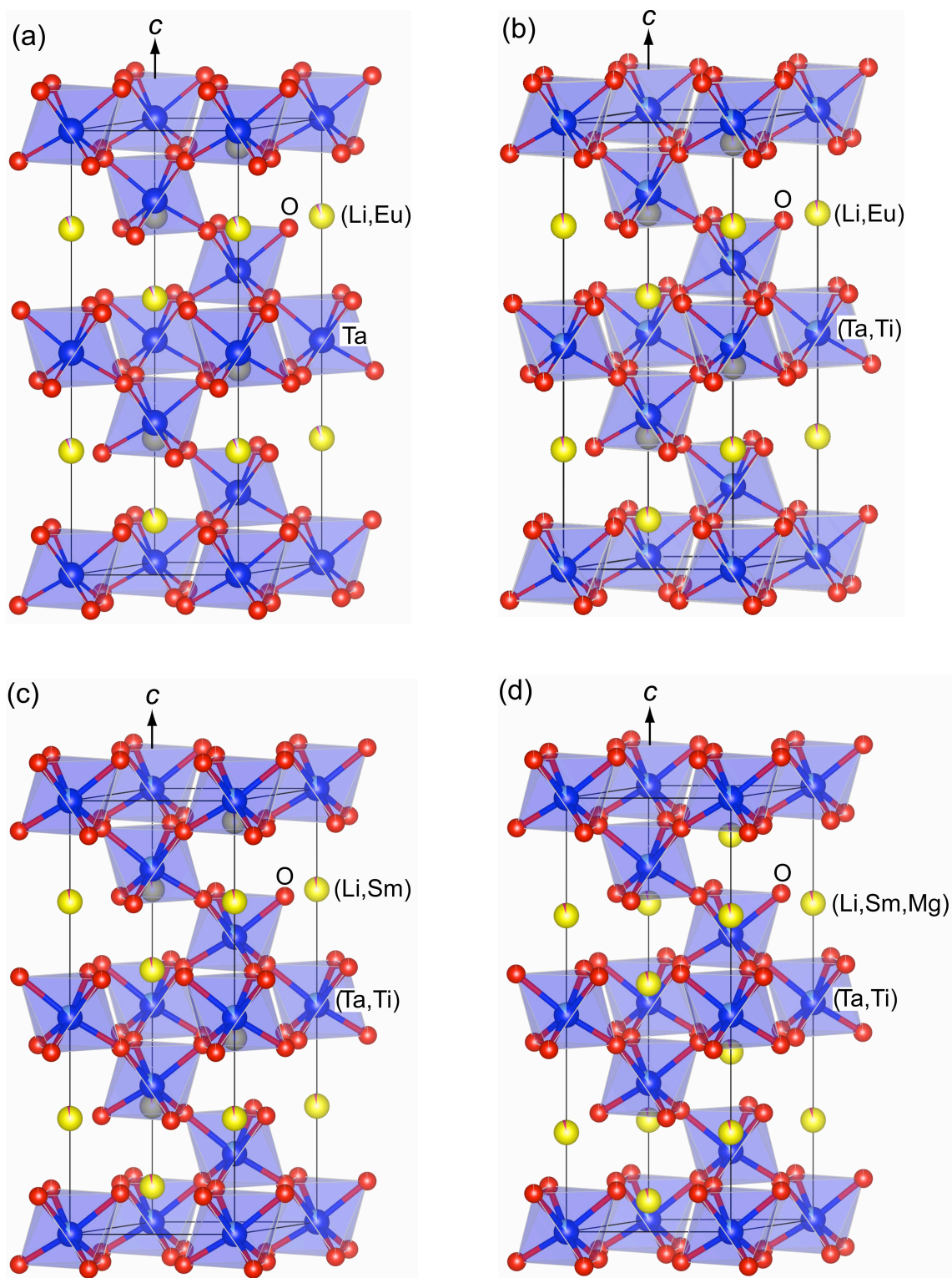
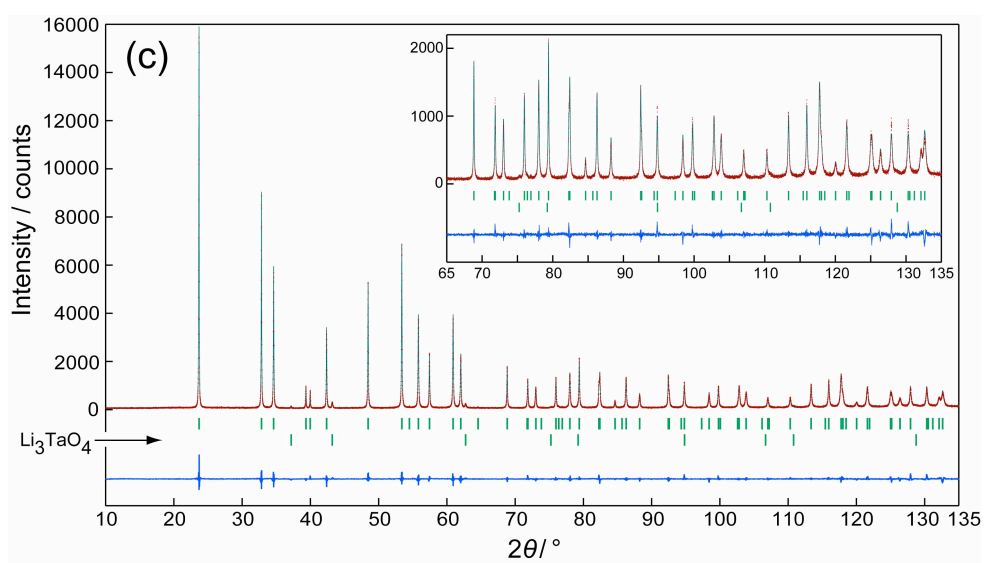
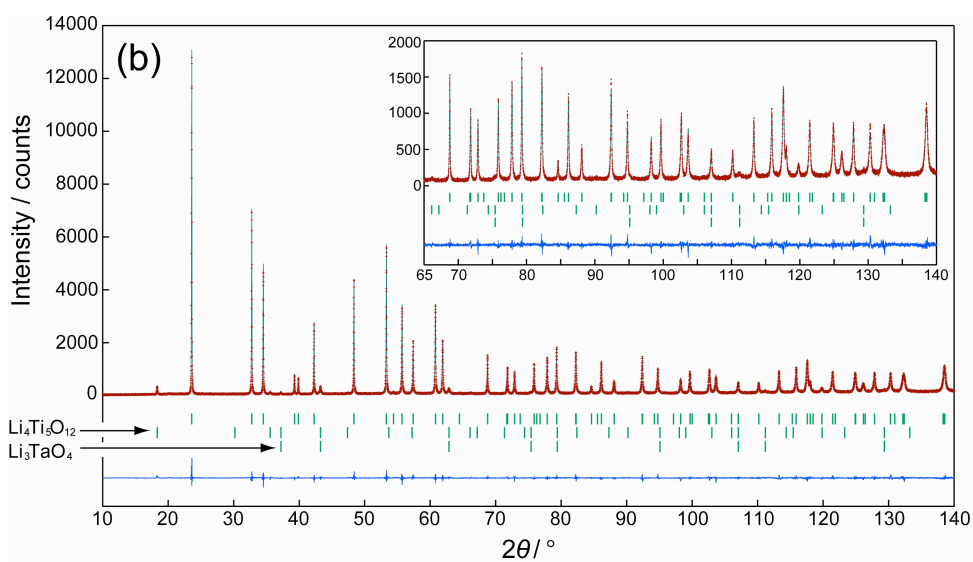
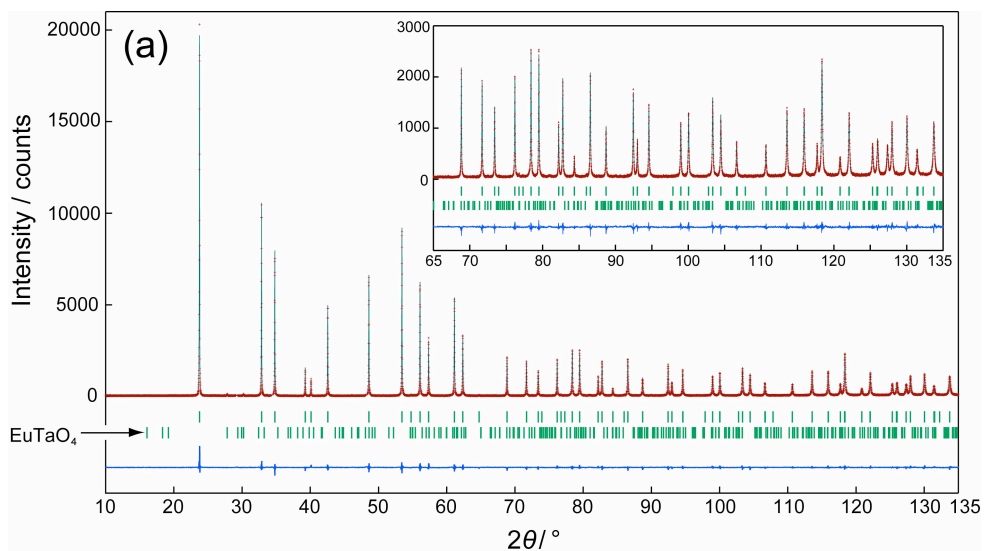
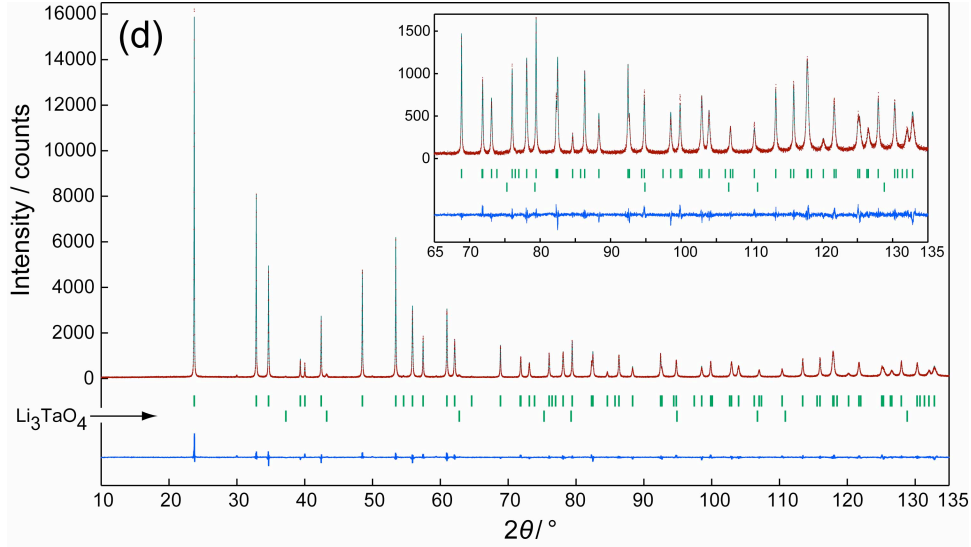


Supplementary data



**Figure S1.** Crystal structures of (a)  $(\text{Li}_{0.925}\text{Eu}^{3+}_{0.025})\text{TaO}_3$  in sample LETO, (b)  $(\text{Li}_{0.968}\text{Eu}^{3+}_{0.032})(\text{Ta}_{0.81}\text{Ti}_{0.19})\text{O}_{2.937}$  in sample LETTO, (c)  $(\text{Li}_{0.967}\text{Sm}^{3+}_{0.033})(\text{Ta}_{0.89}\text{Ti}_{0.11})\text{O}_{2.978}$  in sample LSTTO and (d)  $(\text{Li}_{0.950}\text{Sm}^{3+}_{0.033}\text{Mg}_{0.017})(\text{Ta}_{0.89}\text{Ti}_{0.11})\text{O}_{2.987}$  in sample LSMTTO. Yellow and magenta bicolor balls are for Li (yellow) and (Eu, Sm, Mg) (magenta) sites. Because the occupancy of the oxygen site in (b) is less than unity, the O atoms are displayed as red circle graphs for occupancies. Blue and cyan bicolor balls are for Ta (blue) and Ti (cyan) sites.





**Figure S2.** Comparison of the observed diffraction patterns (symbol: +) with the corresponding calculated patterns (upper solid line). The difference curve is shown in the lower part of each diagram. The vertical bars indicate the positions of possible Bragg reflections. The profile intensities collected for samples (a) LETO, (b) LETTO, (c) LSTTO and (d) LSMTTO.

**TABLE SI.** Reliability indices (%) for Rietveld and MPF methods.

Sample	$R_{wp}$	$S (= R_{wp}/R_e)$	$R_p$	$R_B$	$R_F$
LETO (Rietveld)	10.14	1.29	7.24	2.84	1.40
LETO (MPF)	10.00	1.27	6.89	1.63	0.80
LETTO (Rietveld)	9.06	1.25	6.68	2.19	1.39
LETTO (MPF)	9.01	1.24	6.61	1.49	0.71
LSTTO (Rietveld)	8.88	1.28	6.71	1.96	1.08
LSTTO (MPF)	8.79	1.27	6.62	1.49	0.73
LSMTTO (Rietveld)	9.85	1.29	7.43	2.72	1.23
LSMTTO (MPF)	9.65	1.27	7.17	1.89	0.78

**TABLE SII.** Selected bond lengths (nm) for  $(\text{Li}_{0.925}\text{Eu}^{3+}_{0.025})\text{TaO}_3$  in sample LETO.

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(Li,Eu)–O	0.1987(3)×3
(Li,Eu)–O	0.2648(9)×3
(Li,Eu)–O	0.2964(9)×3
(Li,Eu)–O	0.3191(3)×3
<(Li,Eu)–O>	0.270
Ta–O	0.1905(5)×3
Ta–O	0.2060(5)×3
<Ta–O>	0.198
(Li,Eu)–(Li,Eu)	0.3760(1)
Ta–Ta	0.3760(0)
(Li,Eu)–Ta	0.3037(7)
	0.3069(1)
	0.3358(3)

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**TABLE SIII.** Selected bond lengths (nm) for  $(\text{Li}_{0.968}\text{Eu}^{3+}_{0.032})(\text{Ta}_{0.81}\text{Ti}_{0.19})\text{O}_{2.937}$  in sample LETTO.

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(Li,Eu)–O	0.1992(3)×3
(Li,Eu)–O	0.2767(8)×3
(Li,Eu)–O	0.2833(8)×3
(Li,Eu)–O	0.3195(3)×3
<(Li,Eu)–O>	0.270
(Ta,Ti)–O	0.1930(7)×3
(Ta,Ti)–O	0.2040(6)×3
<(Ta,Ti)–O>	0.199
(Li,Eu)–(Li,Eu)	0.3766(2)
(Ta,Ti)–(Ta,Ti)	0.3766(0)
(Li,Eu)–(Ta,Ti)	0.3122(2)
	0.3178(7)
	0.3302(3)

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**TABLE SIV.** Selected bond lengths (nm) for  $(\text{Li}_{0.967}\text{Sm}^{3+}_{0.033})(\text{Ta}_{0.89}\text{Ti}_{0.11})\text{O}_{2.978}$  in sample LSTTO.

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(Li,Sm)–O	0.2001(4)×3
(Li,Sm)–O	0.2745(1)×3
(Li,Sm)–O	0.2848(9)×3
(Li,Sm)–O	0.3180(3)×3
<(Li,Sm)–O>	0.269
(Ta,Ti)–O	0.1887(8)×3
(Ta,Ti)–O	0.2060(8)×3
<(Ta,Ti)–O>	0.198
(Li,Sm)–(Li,Sm)	0.3765(2)
(Ta,Ti)–(Ta,Ti)	0.3765(0)
(Li,Sm)–(Ta,Ti)	0.3125(2)
	0.3198(6)
	0.3293(2)

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**TABLE SV.** Selected bond lengths (nm) for  $(\text{Li}_{0.950}\text{Sm}^{3+}_{0.033}\text{Mg}_{0.017})(\text{Ta}_{0.89}\text{Ti}_{0.11})\text{O}_{2.987}$  in sample LSMTTO.

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(Li,Sm,Mg)–O	0.2085(4)×3
(Li,Sm,Mg)–O	0.2429(1)×3
(Li,Sm,Mg)–O	0.3176(4)×3
(Li,Sm,Mg)–O	0.3177(1)×3
<(Li,Sm,Mg)–O>	0.272
(Ta,Ti)–O	0.1859(8)×3
(Ta,Ti)–O	0.2076(8)×3
<(Ta,Ti)–O>	0.197
(Li,Sm,Mg)–(Li,Sm,Mg)	0.3764(8)
(Ta,Ti)–(Ta,Ti)	0.3764(0)
(Li,Sm,Mg)–(Ta,Ti)	0.3108(2)
	0.3147(7)
	0.3314(3)

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