

Supplementary information

**Table S1a** Atomic coordinates and isotropic displacement parameters for  $\text{Li}_{3.5}\text{Y}_{1.5}(\text{MoO}_4)_4$  ( $x: 0.00$ ) ( $U_{\text{eq}} = (1/3)\sum_j U_{jj} a_j^* a_j^*$ ).

Atom	Position	x/a	y/b	z/c	s.o.f.	$U_{\text{eq}} / \text{Å}^2$
Li1/Y1	2i	0.12128(9)	0.77267(7)	0.44014(4)	0.25/0.75	0.0109(2)
Mo1	2i	0.33632(6)	0.29999(5)	0.32035(3)		0.0119(1)
Mo2	2i	0.41737(7)	0.15377(6)	0.80009(3)		0.0135(2)
O1	2i	0.1154(6)	0.1162(5)	0.4072(3)		0.0156(5)
O2	2i	0.3013(6)	0.5639(5)	0.5714(3)		0.0178(5)
O3	2i	0.3567(7)	0.1754(5)	0.1622(3)		0.0209(6)
O4	2i	0.1984(6)	0.4994(5)	0.2944(3)		0.0174(5)
O5	2i	0.2703(7)	0.6860(6)	0.0691(3)		0.0252(6)
O6	2i	0.2268(7)	0.3241(5)	0.7720(3)		0.0206(6)
O7	2i	0.8008(7)	0.0584(5)	0.1382(3)		0.0207(6)
O8	2i	0.4850(6)	0.0419(5)	0.6409(3)		0.0170(5)
Li2	1a	0	0	0		0.053(4)
Li3	2i	0.839(3)	0.412(2)	0.121(2)		0.056(3)

**Table S1b** Atomic coordinates and isotropic displacement parameters for  $\text{Li}_{3.5}(\text{Y}_{0.90}\text{Eu}_{0.10})_{1.5}(\text{MoO}_4)_4$  ( $x: 0.10$ ) ( $U_{\text{eq}} = (1/3)\sum_j U_{jj} a_j^* a_j^*$ ).

Atom	Position	x/a	y/b	z/c	s.o.f.	$U_{\text{eq}} / \text{Å}^2$
Li1/Y1/Eu1	2i	0.12138(9)	0.77258(7)	0.44004(4)	0.25/0.675/0.075	0.0083(1)
Mo1	2i	0.33622(7)	0.29991(5)	0.32023(3)		0.0096(1)
Mo2	2i	0.41688(7)	0.15396(6)	0.80032(3)		0.0114(1)
O1	2i	0.1156(6)	0.1167(5)	0.4068(3)		0.0123(5)
O2	2i	0.3017(6)	0.5640(5)	0.5712(3)		0.0152(6)
O3	2i	0.3561(7)	0.1747(5)	0.1624(3)		0.0187(6)
O4	2i	0.1984(7)	0.4993(5)	0.2938(3)		0.0148(6)
O5	2i	0.2702(8)	0.6863(6)	0.0695(3)		0.0231(7)
O6	2i	0.2268(7)	0.3241(5)	0.7725(3)		0.0188(6)
O7	2i	0.8019(7)	0.0584(6)	0.1389(3)		0.0183(6)
O8	2i	0.4854(6)	0.0427(5)	0.6414(3)		0.0155(6)
Li2	1a	0	0	0		0.055(4)
Li3	2i	0.842(3)	0.405(2)	0.124(2)		0.050(3)

**Table S1c** Atomic coordinates and isotropic displacement parameters for  $\text{Li}_{3.5}(\text{Y}_{0.75}\text{Eu}_{0.25})_{1.5}(\text{MoO}_4)_4$  ( $x: 0.25$ ) ( $U_{\text{eq}} = (1/3)\sum_j U_{jj} a_j^* a_j^*$ ).

Atom	Position	x/a	y/b	z/c	s.o.f.	$U_{\text{eq}} / \text{Å}^2$
Li1/Y1/Eu1	2i	0.12066(7)	0.77234(5)	0.43995(3)	0.25/0.5625/0.1875	0.0099(1)
Mo1	2i	0.33560(6)	0.30000(5)	0.31983(3)		0.0111(1)
Mo2	2i	0.41767(7)	0.15410(5)	0.80083(3)		0.0127(1)
O1	2i	0.1164(5)	0.1171(4)	0.4065(3)		0.0143(5)
O2	2i	0.3033(6)	0.5643(4)	0.5717(3)		0.0174(5)
O3	2i	0.3569(6)	0.1756(5)	0.1618(3)		0.0199(5)
O4	2i	0.1962(6)	0.4984(4)	0.2936(3)		0.0168(5)
O5	2i	0.2686(7)	0.6866(5)	0.0686(3)		0.0249(6)
O6	2i	0.2287(6)	0.3251(5)	0.7733(3)		0.0205(6)
O7	2i	0.8011(6)	0.0575(5)	0.1378(3)		0.0198(5)
O8	2i	0.4859(6)	0.0435(4)	0.6420(3)		0.0166(5)
Li2	1a	0	0	0		0.059(4)
Li3	2i	0.841(3)	0.404(2)	0.125(2)		0.053(3)

**Table S1d** Atomic coordinates and isotropic displacement parameters for  $\text{Li}_{3.5}(\text{Y}_{0.50}\text{Eu}_{0.50})_{1.5}(\text{MoO}_4)_4$  ( $x: 0.50$ ) ( $U_{\text{eq}} = (1/3)\sum_j U_{jj} a_j^* a_j$ ).

Atom	Position	$x/a$	$y/b$	$z/c$	s.o.f.	$U_{\text{eq}} / \text{\AA}^2$
Li1/Eu1/Y1	2i	0.12033(7)	0.77205(6)	0.43980(3)	0.25/0.375/0.375	0.0085(1)
Mo1	2i	0.33484(7)	0.29990(6)	0.31928(4)		0.0103(1)
Mo2	2i	0.41777(8)	0.15446(7)	0.80161(4)		0.0120(1)
O1	2i	0.1184(6)	0.1175(5)	0.4056(3)		0.0126(6)
O2	2i	0.3048(6)	0.5650(5)	0.5721(3)		0.0157(6)
O3	2i	0.3557(7)	0.1757(6)	0.1620(3)		0.0179(7)
O4	2i	0.1945(7)	0.4965(5)	0.2931(3)		0.0149(6)
O5	2i	0.2685(8)	0.6877(6)	0.0687(3)		0.0249(8)
O6	2i	0.2313(7)	0.3252(6)	0.7739(3)		0.0200(7)
O7	2i	0.8003(7)	0.0563(6)	0.1378(3)		0.0183(7)
O8	2i	0.4837(6)	0.0439(5)	0.6429(3)		0.0159(6)
Li2	1a	0	0	0		0.053(5)
Li3	2i	0.831(3)	0.397(3)	0.125(2)		0.061(4)

**Table S1e** Atomic coordinates and isotropic displacement parameters for  $\text{Li}_{3.5}(\text{Y}_{0.25}\text{Eu}_{0.75})_{1.5}(\text{MoO}_4)_4$  ( $x: 0.75$ ) ( $U_{\text{eq}} = (1/3)\sum_j U_{jj} a_j^* a_j$ ).

Atom	Position	$x/a$	$y/b$	$z/c$	s.o.f.	$U_{\text{eq}} / \text{\AA}^2$
Li1/Y1/Eu1	2i	0.12001(6)	0.77183(5)	0.43964(3)	0.25/0.1875/0.5625	0.0094(1)
Mo1	2i	0.33420(7)	0.29986(5)	0.31873(3)		0.0106(1)
Mo2	2i	0.41808(8)	0.15474(6)	0.80227(3)		0.0123(1)
O1	2i	0.1196(6)	0.1175(5)	0.4049(3)		0.0129(6)
O2	2i	0.3063(7)	0.5643(5)	0.5725(3)		0.0158(6)
O3	2i	0.3562(7)	0.1753(6)	0.1615(3)		0.0192(6)
O4	2i	0.1922(7)	0.4953(5)	0.2921(3)		0.0166(6)
O5	2i	0.2683(8)	0.6894(6)	0.0691(3)		0.0241(7)
O6	2i	0.2320(7)	0.3259(5)	0.7752(3)		0.0193(6)
O7	2i	0.7997(7)	0.0556(6)	0.1368(3)		0.0194(6)
O8	2i	0.4826(7)	0.0445(5)	0.6429(3)		0.0159(6)
Li2	1a	0	0	0		0.064(5)
Li3	2i	0.839(3)	0.403(2)	0.124(2)		0.053(3)

**Table S1f** Atomic coordinates and isotropic displacement parameters for  $\text{Li}_{3.5}\text{Eu}_{1.5}(\text{MoO}_4)_4$  ( $x: 1.00$ ) ( $U_{\text{eq}} = (1/3)\sum_j U_{jj} a_j^* a_j$ ).

Atom	Position	$x/a$	$y/b$	$z/c$	s.o.f.	$U_{\text{eq}} / \text{\AA}^2$
Li1/Eu1	2i	0.11989(4)	0.77156(4)	0.43946(2)	0.25/0.75	0.0095(1)
Mo1	2i	0.33392(6)	0.29982(5)	0.31824(3)		0.0104(1)
Mo2	2i	0.41798(7)	0.15515(6)	0.80287(3)		0.0121(1)
O1	2i	0.1204(5)	0.1170(5)	0.4046(3)		0.0135(5)
O2	2i	0.3074(5)	0.5645(5)	0.5730(3)		0.0170(6)
O3	2i	0.3555(6)	0.1745(5)	0.1615(3)		0.0196(6)
O4	2i	0.1912(6)	0.4943(5)	0.2915(3)		0.0161(6)
O5	2i	0.2689(6)	0.6893(6)	0.0687(3)		0.0231(7)
O6	2i	0.2333(6)	0.3266(5)	0.7764(3)		0.0188(6)
O7	2i	0.7991(6)	0.0555(5)	0.1366(3)		0.0193(6)
O8	2i	0.4810(5)	0.0453(5)	0.6435(3)		0.0159(6)
Li2	1a	0	0	0		0.045(3)
Li3	2i	0.829(3)	0.403(3)	0.120(2)		0.063(3)

**Table S2a** Anisotropic displacement parameters of  $\text{Li}_{3.5}\text{Y}_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.00).

Atom	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Li1/Y1	0.0106(3)	0.0122(3)	0.0110(3)	0.0056(2)	0.0037(2)	0.0033(2)
Mo1	0.01103(19)	0.0139(2)	0.01217(19)	0.00595(14)	0.00398(13)	0.00431(14)
Mo2	0.0142(2)	0.0175(2)	0.01087(19)	0.00847(14)	0.00439(12)	0.00308(13)
O1	0.0183(13)	0.0149(12)	0.0166(12)	0.0090(11)	0.0068(10)	0.0046(10)
O2	0.0154(13)	0.0182(13)	0.0197(13)	0.0072(11)	0.0035(11)	0.0054(11)
O3	0.0185(14)	0.0248(15)	0.0190(13)	0.0112(12)	0.0045(11)	0.0008(11)
O4	0.0214(14)	0.0183(13)	0.0173(12)	0.0126(12)	0.0064(11)	0.0051(11)
O5	0.0226(16)	0.0293(17)	0.0178(13)	0.0061(13)	0.0019(12)	0.0062(12)
O6	0.0220(15)	0.0199(14)	0.0237(13)	0.0120(12)	0.0073(12)	0.0066(12)
O7	0.0231(15)	0.0246(15)	0.0201(13)	0.0129(13)	0.0095(11)	0.0092(12)
O8	0.0171(13)	0.0176(13)	0.0168(12)	0.0076(11)	0.0076(10)	0.0019(10)

**Table S2b** Anisotropic displacement parameters of  $\text{Li}_{3.5}(\text{Y}_{0.90}\text{Eu}_{0.10})_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.10).

Atom	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Li1/Y1/Eu1	0.0088(2)	0.0071(2)	0.0097(2)	0.00359(17)	0.00319(15)	0.00211(16)
Mo1	0.00886(18)	0.00936(18)	0.01106(18)	0.00365(13)	0.00355(12)	0.00359(12)
Mo2	0.01222(19)	0.01321(19)	0.00962(18)	0.00631(14)	0.00366(12)	0.00235(12)
O1	0.0120(13)	0.0115(13)	0.0140(12)	0.0047(11)	0.005(1)	0.0039(10)
O2	0.0138(14)	0.0126(13)	0.0168(13)	0.0032(11)	0.0038(11)	0.0037(11)
O3	0.0163(14)	0.0204(15)	0.0183(14)	0.0089(12)	0.0044(11)	-0.0006(11)
O4	0.0204(15)	0.0120(13)	0.0159(13)	0.0097(12)	0.0080(11)	0.0032(10)
O5	0.0228(17)	0.0246(17)	0.0132(14)	0.0042(14)	-0.0002(12)	0.0022(12)
O6	0.0217(16)	0.0158(14)	0.0210(15)	0.0106(13)	0.0073(12)	0.0023(12)
O7	0.0178(15)	0.0213(15)	0.0204(14)	0.0101(13)	0.0085(11)	0.0092(12)
O8	0.0148(14)	0.0135(13)	0.0167(13)	0.0046(11)	0.0068(11)	0.0012(11)

**Table S2c** Anisotropic displacement parameters of  $\text{Li}_{3.5}(\text{Y}_{0.75}\text{Eu}_{0.25})_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.25).

Atom	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Li1/Y1/Eu1	0.0111(2)	0.0101(2)	0.0102(2)	0.00552(16)	0.00327(15)	0.00362(16)
Mo1	0.01137(18)	0.01234(19)	0.01146(19)	0.00587(13)	0.00377(12)	0.00499(13)
Mo2	0.01446(19)	0.01572(19)	0.00996(18)	0.00821(13)	0.00377(12)	0.00361(12)
O1	0.0166(12)	0.0145(11)	0.0146(12)	0.0094(10)	0.0042(9)	0.0044(9)
O2	0.0160(13)	0.0173(12)	0.0171(13)	0.0055(10)	0.0028(10)	0.0052(10)
O3	0.0187(13)	0.0237(13)	0.0167(13)	0.0108(11)	0.0038(10)	0.0008(10)
O4	0.0208(13)	0.0164(11)	0.0173(13)	0.0111(10)	0.0064(10)	0.0051(9)
O5	0.0243(15)	0.0260(14)	0.0161(14)	0.0039(12)	0.0018(11)	0.0038(11)
O6	0.0233(14)	0.0193(12)	0.0230(14)	0.0123(11)	0.0084(11)	0.0058(11)
O7	0.0215(14)	0.0249(13)	0.0192(13)	0.0132(11)	0.0091(11)	0.0095(11)
O8	0.0154(12)	0.0174(11)	0.0166(12)	0.0057(10)	0.0071(10)	0.0035(10)

**Table S2d** Anisotropic displacement parameters of  $\text{Li}_{3.5}(\text{Y}_{0.50}\text{Eu}_{0.50})_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.50).

Atom	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Li1/Y1/Eu1	0.00842(19)	0.0074(2)	0.0088(2)	0.00290(15)	0.00219(14)	0.00092(15)
Mo1	0.00935(19)	0.0106(2)	0.0105(2)	0.00381(15)	0.00283(14)	0.00272(15)
Mo2	0.0124(2)	0.0142(2)	0.00903(19)	0.00601(15)	0.00306(14)	0.00147(14)
O1	0.0123(13)	0.0089(14)	0.0174(15)	0.0040(12)	0.0060(11)	0.0044(12)
O2	0.0129(14)	0.0148(16)	0.0153(14)	0.0024(12)	0.0022(11)	0.0030(12)
O3	0.0164(14)	0.0223(17)	0.0140(15)	0.0096(13)	0.0044(12)	-0.0013(13)
O4	0.0186(14)	0.0114(14)	0.0151(14)	0.0073(12)	0.0041(12)	0.0021(12)
O5	0.0234(16)	0.0257(19)	0.0155(16)	0.0019(15)	0.0015(13)	0.0040(14)
O6	0.0211(16)	0.0192(17)	0.0218(17)	0.0120(14)	0.0060(13)	0.0025(14)
O7	0.0167(15)	0.0224(18)	0.0181(15)	0.0078(13)	0.0078(12)	0.0083(14)
O8	0.0150(14)	0.0140(15)	0.0173(15)	0.0047(13)	0.0060(12)	0.0021(12)

**Table S2e** Anisotropic displacement parameters of  $\text{Li}_{3.5}(\text{Y}_{0.25}\text{Eu}_{0.75})_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.75).

Atom	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Li1/Y1/Eu1	0.01071(17)	0.00821(16)	0.00922(15)	0.00370(12)	0.00345(11)	0.00214(11)
Mo1	0.01118(19)	0.01089(19)	0.01052(17)	0.00443(15)	0.00403(13)	0.00367(13)
Mo2	0.01411(19)	0.01454(19)	0.00907(16)	0.00657(15)	0.00402(13)	0.00254(12)
O1	0.0133(13)	0.0119(14)	0.0158(12)	0.0056(11)	0.0067(11)	0.0052(10)
O2	0.0155(15)	0.0130(14)	0.0162(13)	0.0030(12)	0.0039(11)	0.0044(11)
O3	0.0170(15)	0.0227(17)	0.0162(13)	0.0093(14)	0.0036(11)	-0.0002(12)
O4	0.0197(15)	0.0152(14)	0.0155(13)	0.0084(13)	0.0042(11)	0.0041(11)
O5	0.0221(17)	0.0264(18)	0.0142(13)	0.0028(15)	0.0016(12)	0.0023(12)
O6	0.0218(16)	0.0168(16)	0.0205(14)	0.0095(14)	0.0065(12)	0.0036(12)
O7	0.0207(16)	0.0252(17)	0.0180(13)	0.0116(14)	0.0095(12)	0.0102(12)
O8	0.0154(15)	0.0156(15)	0.0150(13)	0.0038(12)	0.0067(11)	0.0030(11)

**Table S2f** Anisotropic displacement parameters of  $\text{Li}_{3.5}(\text{Eu}_{1.5}(\text{MoO}_4)_4)$  ( $x$ : 1.00).

Atom	$U_{11}$	$U_{22}$	$U_{33}$	$U_{12}$	$U_{13}$	$U_{23}$
Eu1/Eu1	0.00999(13)	0.00818(14)	0.01075(14)	0.00354(10)	0.00375(10)	0.0028(1)
Mo1	0.01014(16)	0.01072(19)	0.01162(17)	0.00447(13)	0.00437(13)	0.00458(14)
Mo2	0.01302(16)	0.01416(19)	0.01011(17)	0.00632(13)	0.00420(12)	0.00321(13)
O1	0.0135(10)	0.0109(13)	0.0183(13)	0.0053(10)	0.0076(10)	0.0052(11)
O2	0.0157(11)	0.0155(15)	0.0177(14)	0.0036(10)	0.0042(10)	0.0063(11)
O3	0.0173(12)	0.0234(16)	0.0184(14)	0.0102(12)	0.0064(10)	0.0007(12)
O4	0.0210(12)	0.0146(14)	0.0170(14)	0.0103(11)	0.0072(10)	0.0062(11)
O5	0.0224(13)	0.0247(17)	0.0157(14)	0.0043(12)	0.0022(11)	0.0054(13)
O6	0.0206(13)	0.0184(16)	0.0198(15)	0.0110(11)	0.0056(11)	0.0047(12)
O7	0.0200(13)	0.0234(16)	0.0206(14)	0.0112(12)	0.0102(11)	0.0107(13)
O8	0.0135(11)	0.0156(14)	0.0179(14)	0.0043(10)	0.0071(10)	0.0041(11)

**Table S3a** Interatomic distances in  $\text{Li}_{3.5}(\text{Y}_{1-x}\text{Eu}_x)_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.00, 0.10, 0.25, 0.50, 0.75, 1.00) in the  $[\text{M1O}_8]$ -polyhedra ( $\text{M1}$ : 75%  $(\text{Y}_{1-x}\text{Eu}_x)$  + 25% Li) in Å.

$d(\text{M1},\text{O}_x)$	Sym. $\text{O}_x$	$x$ : 0.00	$x$ : 0.10	$x$ : 0.25	$x$ : 0.50	$x$ : 0.75	$x$ : 1.00
$d(\text{M1},\text{O1})$	$x, 1+y, z$	2.374(3)	2.382(3)	2.389(2)	2.400(3)	2.407(3)	2.417(3)
$d(\text{M1},\text{O1})$	$-x, 1-y, 1-z$	2.338(3)	2.345(3)	2.350(2)	2.370(3)	2.384(3)	2.395(3)
$d(\text{M1},\text{O2})$	$x, y, z$	2.405(3)	2.407(3)	2.417(3)	2.425(3)	2.441(3)	2.452(3)
$d(\text{M1},\text{O2})$	$-x, 1-y, 1-z$	2.442(3)	2.449(3)	2.454(3)	2.467(3)	2.473(3)	2.482(3)
$d(\text{M1},\text{O4})$	$x, y, z$	2.345(3)	2.350(3)	2.352(2)	2.364(3)	2.373(3)	2.383(3)
$d(\text{M1},\text{O6})$	$-x, 1-y, 1-z$	2.380(3)	2.385(3)	2.393(3)	2.405(3)	2.417(3)	2.430(3)
$d(\text{M1},\text{O8})$	$x, 1+y, z$	2.411(3)	2.419(3)	2.429(3)	2.438(3)	2.442(3)	2.453(3)
$d(\text{M1},\text{O8})$	$1-x, 1-y, 1-z$	2.338(3)	2.341(3)	2.347(3)	2.367(3)	2.376(3)	2.391(3)

**Table S3b** Interatomic distances in  $\text{Li}_{3.5}(\text{Y}_{1-x}\text{Eu}_x)_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.00, 0.10, 0.25, 0.50, 0.75, 1.00) in the  $[\text{Mo1O}_4]$ -tetrahedra in Å.

$d(\text{Mo1},\text{O}_x)$	Sym. $\text{O}_x$	$x$ : 0.00	$x$ : 0.10	$x$ : 0.25	$x$ : 0.50	$x$ : 0.75	$x$ : 1.00
$d(\text{Mo1},\text{O1})$	$x, y, z$	1.816(3)	1.814(3)	1.813(3)	1.807(3)	1.806(3)	1.811(3)
$d(\text{Mo1},\text{O2})$	$1-x, 1-y, 1-z$	1.795(3)	1.797(3)	1.794(3)	1.795(3)	1.794(3)	1.792(3)
$d(\text{Mo1},\text{O3})$	$x, y, z$	1.729(3)	1.729(3)	1.733(3)	1.728(3)	1.733(3)	1.730(3)
$d(\text{Mo1},\text{O4})$	$x, y, z$	1.760(3)	1.764(3)	1.765(2)	1.762(3)	1.766(3)	1.769(3)

**Table S3c** Interatomic distances in  $\text{Li}_{3.5}(\text{Y}_{1-x}\text{Eu}_x)_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.00, 0.10, 0.25, 0.50, 0.75, 1.00) in the  $[\text{Mo2O}_4]$ -tetrahedra in Å.

$d(\text{Mo2},\text{O}_x)$	Sym. $\text{O}_x$	$x$ : 0.00	$x$ : 0.10	$x$ : 0.25	$x$ : 0.50	$x$ : 0.75	$x$ : 1.00
$d(\text{Mo2},\text{O5})$	$1-x, 1-y, 1-z$	1.732(3)	1.732(3)	1.737(3)	1.733(4)	1.728(3)	1.724(3)
$d(\text{Mo2},\text{O6})$	$x, y, z$	1.785(3)	1.784(3)	1.787(3)	1.781(3)	1.784(3)	1.784(3)
$d(\text{Mo2},\text{O7})$	$1-x, -y, 1-z$	1.744(3)	1.744(3)	1.747(3)	1.743(3)	1.747(3)	1.749(3)
$d(\text{Mo2},\text{O8})$	$x, y, z$	1.823(3)	1.823(3)	1.822(3)	1.818(3)	1.822(3)	1.818(3)

**Table S3d** Interatomic distances in  $\text{Li}_{3.5}(\text{Y}_{1-x}\text{Eu}_x)_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.00, 0.10, 0.25, 0.50, 0.75, 1.00) in the  $[\text{Li2O}_4]$ -polyhedra in Å.

$d(\text{Li2},\text{O}_x)$	Sym. $\text{O}_x$	$x$ : 0.00	$x$ : 0.10	$x$ : 0.25	$x$ : 0.50	$x$ : 0.75	$x$ : 1.00
$d(\text{Li2},\text{O3})$	$x, y, z$	2.042(3)	2.043(3)	2.043(3)	2.044(3)	2.044(3)	2.046(3)
$d(\text{Li2},\text{O3})$	$-x, -y, -z$	2.042(3)	2.043(3)	2.043(3)	2.044(3)	2.044(3)	2.046(3)
$d(\text{Li2},\text{O7})$	$1-x, -y, -z$	1.966(3)	1.969(3)	1.963(3)	1.969(3)	1.965(3)	1.970(3)
$d(\text{Li2},\text{O7})$	$-1+x, y, z$	1.966(3)	1.969(3)	1.963(3)	1.969(3)	1.965(3)	1.970(3)
$d(\text{Li2},\text{O5})$	$x, -1+y, z$	3.027(5)	3.031(5)	3.025(4)	3.026(5)	3.023(5)	3.034(5)
$d(\text{Li2},\text{O5})$	$-x, 1-y, -z$	3.027(5)	3.031(5)	3.025(4)	3.026(5)	3.023(5)	3.034(5)

**Table S3e** Interatomic distances in  $\text{Li}_{3.5}(\text{Y}_{1-x}\text{Eu}_x)_{1.5}(\text{MoO}_4)_4$  ( $x$ : 0.00, 0.10, 0.25, 0.50, 0.75, 1.00) in the  $[\text{Li3O}_6]$ -octahedra in Å.

$d(\text{Li3},\text{O}_x)$	Sym. $\text{O}_x$	$x$ : 0.00	$x$ : 0.10	$x$ : 0.25	$x$ : 0.50	$x$ : 0.75	$x$ : 1.00
$d(\text{Li3},\text{O3})$	$x, y, z$	2.566(15)	2.557(14)	2.548(13)	2.506(16)	2.555(15)	2.606(16)
$d(\text{Li3},\text{O4})$	$1+x, y, z$	2.151(15)	2.112(14)	2.102(14)	2.127(17)	2.104(14)	2.085(17)
$d(\text{Li3},\text{O5})$	$1+x, y, z$	2.513(15)	2.544(14)	2.555(14)	2.622(16)	2.580(15)	2.543(16)
$d(\text{Li3},\text{O5})$	$1-x, 1-y, -z$	1.855(15)	1.892(14)	1.890(15)	1.889(18)	1.884(14)	1.894(17)
$d(\text{Li3},\text{O6})$	$1-x, 1-y, 1-z$	2.077(15)	2.105(14)	2.108(13)	2.131(17)	2.122(15)	2.079(16)
$d(\text{Li3},\text{O7})$	$x, y, z$	2.319(15)	2.273(14)	2.271(13)	2.252(17)	2.293(14)	2.365(15)

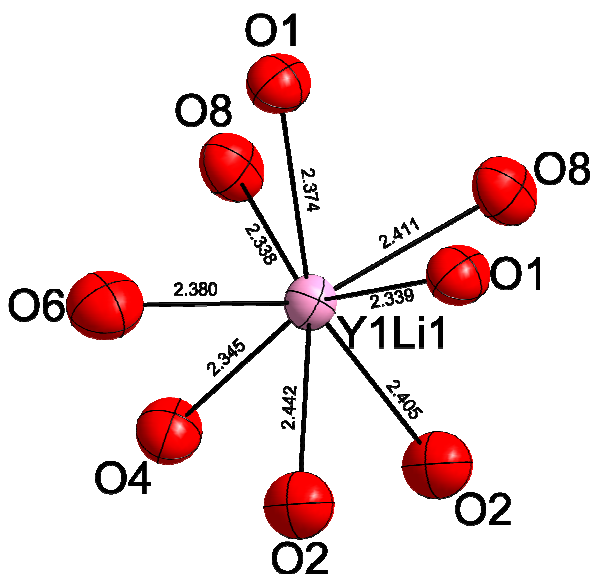


Fig. S1 ORTEP style representation of  $M1O_8$ -coordination polyhedra ( $M1$ : 75%Y + 25%Li) in  $Li_{3.5}Y_{1.5}(MoO_4)_4$ . Ellipsoids at 90% probability level.

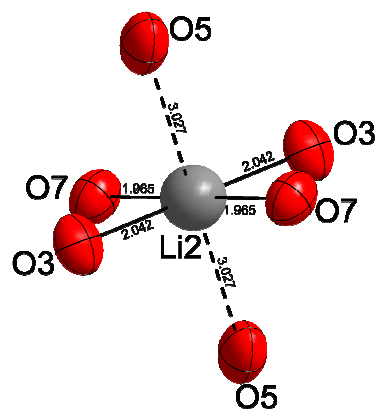


Fig. S4 ORTEP style representation of  $Li_2O_4$ -coordination polyhedra in  $Li_{3.5}Y_{1.5}(MoO_4)_4$ . Ellipsoids at 90% probability level.

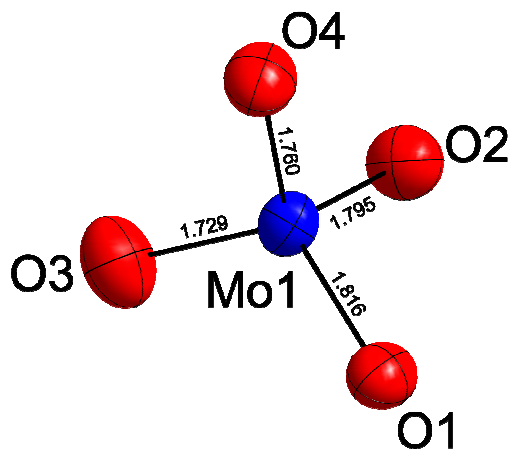


Fig. S2 ORTEP style representation of  $Mo_1O_4$ -coordination polyhedra in  $Li_{3.5}Y_{1.5}(MoO_4)_4$ . Ellipsoids at 90% probability level.

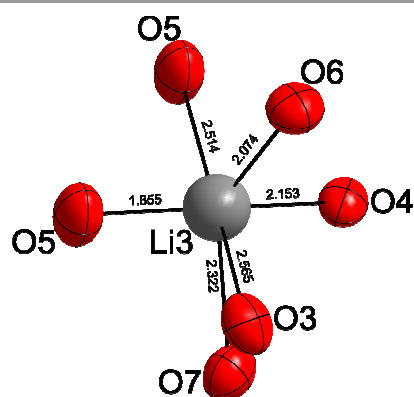


Fig. S5 ORTEP style representation of  $Li_3O_4$ -coordination polyhedra in  $Li_{3.5}Y_{1.5}(MoO_4)_4$ . Ellipsoids at 90% probability level.

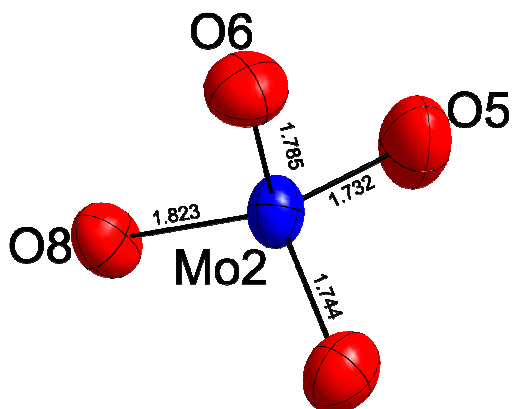


Fig. S3 ORTEP style representation of  $Mo_2O_4$ -coordination polyhedra in  $Li_{3.5}Y_{1.5}(MoO_4)_4$ . Ellipsoids at 90% probability level.