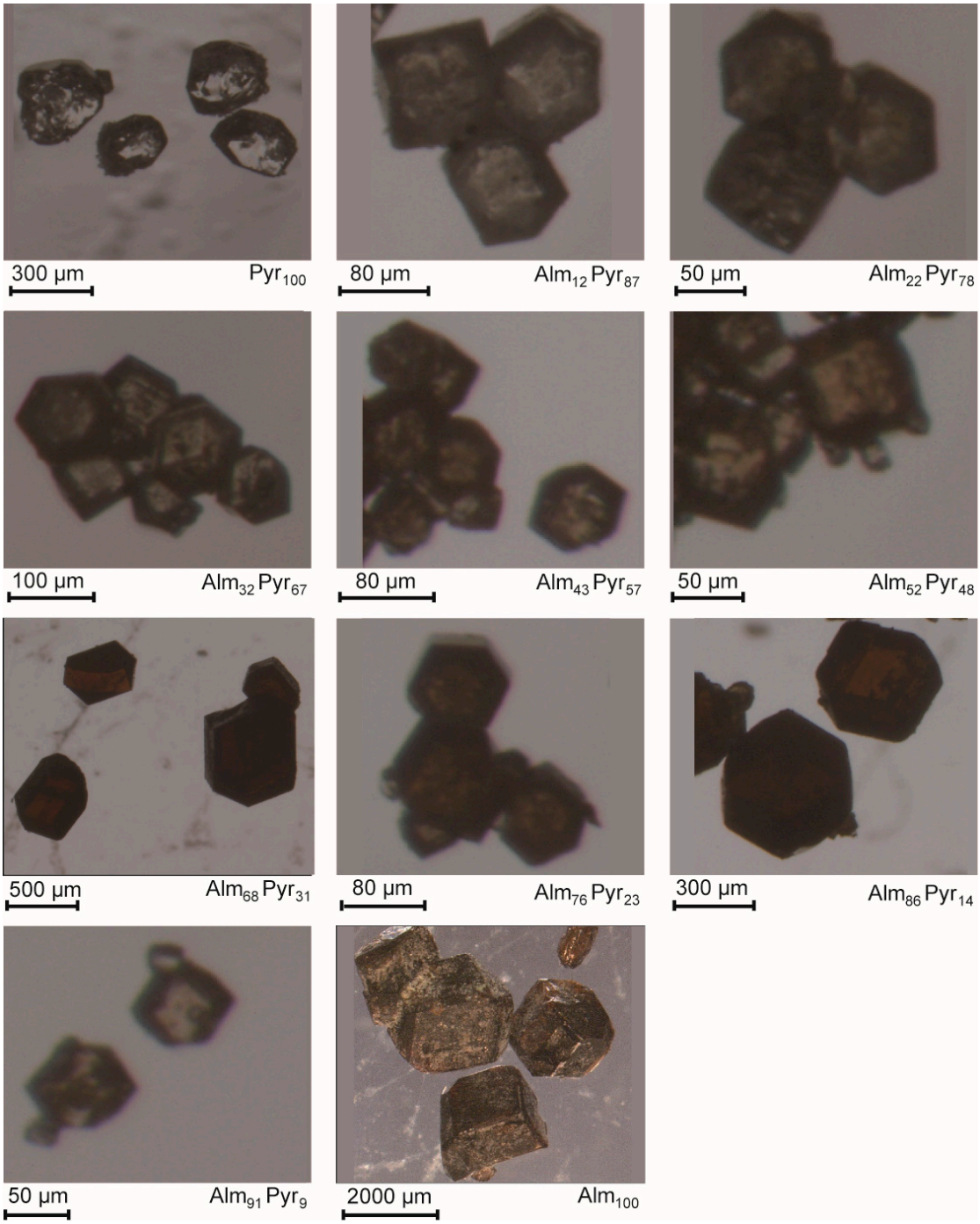

Figure S1. Microphotographs of the synthetic Alm-Pyr crystals.

Table S1. Details of the structural refinement of synthesis single garnet crystals.

(1) Alm_{100}

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$Fe_{3.13}Al_{1.9}Ti_{0.1}Si_{2.91}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.53025(9)Å, α=β=γ=90°
Volume ($Å^3$)	1532.91(4)

R_{int}	0.0536
All reflections	5981
Great reflections	133
Refined parameters	17
R_1	0.0197
wR_2	0.0558
S	1.1704

Atom	Occupancy	x	y	z	U_{iso}
Fe	1	0.875	0.5	0.25	0.0062(3)
Si	1	0.125	0.5	0.25	0.0021(4)
Al	1	0	0.5	0.5	0.0017(4)
O	1	0.03415(8)	0.54921(8)	0.34725(8)	0.0035(4)

$[AlO_6]$ octahedron
Average bond length = 1.8918(10) Å
Octahedron volume = 9.0206 $Å^3$
Effective coordination number = 6
$[FeO_8]$ dodecahedron
Average bond length = 2.2971(10) Å
bond I length = 2.2241(9) Å
bond II length = 2.3701(10) Å
Polyhedral volume = 20.8362 $Å^3$
Effective coordination number = 7.6815
$[SiO_4]$ tetrahedron
Average bond length = 1.6360(10) Å
Polyhedral volume = 2.2058 $Å^3$
Effective coordination number = 4

(2) Pyr_9Alm_{91}

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$(Fe_{2.88}Mg_{0.27})\ Al_{1.89}Ti_{0.05}Si_{2.94}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.52701(11)Å, α=β=γ=90°
Volume ($Å^3$)	1531.62(4)
R_{int}	0.0369
All reflections	5985
Great reflections	123
Refined parameters	19
R_1	0.0181
wR_2	0.0511
S	1.0883

Atom	Occupancy	x	y	z	U_{iso}
Mg	0.11(2)	0.875	0.5	0.25	0.0063(4)
Fe	0.899(11)	0.875	0.5	0.25	0.0063(4)
Si	1	0.125	0.5	0.25	0.0023(5)
Al	1	0	0.5	0.5	0.0019(5)
O	1	0.03402(8)	0.54910(9)	0.34724(8)	0.0048(5)

$[AlO_6]$ octahedron
Average bond length = 1.8907(11) Å
Octahedron volume = 9.0044 $Å^3$
Effective coordination number = 6
$[FeO_8]$ /$[MgO_8]$ dodecahedron
Average bond length = 2.2962(10) Å

bond I length = 2.2218(10) Å
bond II length = 2.3704(10) Å
Polyhedral volume = 20.8197 Å^3
Effective coordination number =7.6691
[SiO_4] tetrahedron
Average bond length = 1.6360(10) Å
Polyhedral volume = 2.2047 Å^3
Effective coordination number = 4

(3) $Pyr_{14}Alm_{86}$

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$(Fe_{2.71}Mg_{0.43})\ Al_{1.92}Ti_{0.07}Si_{2.92}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.5241(3)Å, α=β=γ=90°
Volume (Å^3)	1530.46(12)
R_{int}	0.0632
All reflections	5656
Great reflections	108
Refined parameters	19
R_1	0.0225
wR_2	0.0568
S	1.1715

Atom	Occupancy	x	y	z	U_{iso}
Mg	0.16(2)	0.875	0.5	0.25	0.0057(5)
Fe	0.842(13)	0.875	0.5	0.25	0.0057(5)
Si	1	0.125	0.5	0.25	0.0020(7)
Al	1	0	0.5	0.5	0.0011(7)
O	1	0.0339(11)	0.54928(10)	0.34724(10)	0.0019(6)

[AlO_6] octahedron
Average bond length = 1.8931(13) Å
Octahedron volume = 9.0381 Å^3
Effective coordination number = 6
[FeO_8] /[MgO_8] dodecahedron
Average bond length = 2.2936(12) Å
bond I length = 2.2193(12) Å
bond II length = 2.3680(12) Å
Polyhedral volume = 20.7538 Å^3
Effective coordination number =7.6679
[SiO_4] tetrahedron
Average bond length = 1.6353(13) Å
Polyhedral volume = 2.2006 Å^3
Effective coordination number = 4

(4) $Pyr_{23}Alm_{76}$

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$(Fe_{2.55}Mg_{0.78})\ Al_{1.87}Ca_{0.01}Ti_{0.05}Si_{2.88}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.52759(16)Å, α=β=γ=90°
Volume (Å^3)	1531.85(6)
R_{int}	0.0382

All reflections			5797		
Great reflections			116		
Refined parameters			19		
R_1			0.0171		
wR_2			0.0484		
S			1.1512		
Atom	Occupancy	x	y	z	U_{iso}
Mg	0.28(2)	0.875	0.5	0.25	0.0110(4)
Fe	0.729(10)	0.875	0.5	0.25	0.0110(4)
Si	1	0.125	0.5	0.25	0.0075(4)
Al	1	0	0.5	0.5	0.0043(5)
O	1	0.03391(8)	0.54951(8)	0.34692(8)	0.0100(4)

$[AlO_6]$ octahedron
Average bond length = 1.8954(10) Å
Octahedron volume = 9.0697 Å^3
Effective coordination number = 6
$[FeO_8]$ /$[MgO_8]$ dodecahedron
Average bond length = 2.2933(9) Å
bond I length = 2.2203(9) Å
bond II length = 2.3662(9) Å
Polyhedral volume = 20.7357 Å^3
Effective coordination number =7.6806
$[SiO_4]$ tetrahedron
Average bond length = 1.6360(10) Å
Polyhedral volume = 2.2037 Å^3
Effective coordination number = 4

(5) $Pyr_{31}Alm_{68}$

Wavelength (Å)			0.4340		
Pressure (GPa)			0.0001		
Temperature (K)			298		
Composition			$(Fe_{2.16}Mg_{0.99})$ $Al_{1.9}Ca_{0.01}Ti_{0.08}Si_{2.92}O_{12}$		
Symmetry			cubic, $Ia\bar{3}d$		
Lattice parameters			a=b=c=11.51055(6)Å, α=β=γ=90°		
Volume (Å^3)			1525.06(2)		
R_{int}			0.0635		
All reflections			5851		
Great reflections			106		
Refined parameters			19		
R_1			0.0233		
wR_2			0.0607		
S			1.1479		
Atom	Occupancy	x	y	z	U_{iso}
Mg	0.41(2)	0.875	0.5	0.25	0.0080(5)
Fe	0.600(10)	0.875	0.5	0.25	0.0080(5)
Si	1	0.125	0.5	0.25	0.0046(5)
Al	1	0	0.5	0.5	0.0033(6)
O	1	0.0337(8)	0.54967(8)	0.34694(8)	0.0054(5)

$[AlO_6]$ octahedron
Average bond length = 1.8925(10) Å
Octahedron volume = 9.0269 Å^3
Effective coordination number = 6
$[FeO_8]$ /$[MgO_8]$ dodecahedron
Average bond length = 2.2882(9) Å

bond I length = 2.2159(9) Å
bond II length = 2.3606(9) Å
Polyhedral volume = 20.5985 Å^3
Effective coordination number =7.685
[SiO_4] tetrahedron
Average bond length = 1.6357(10) Å
Polyhedral volume = 2.2019 Å^3
Effective coordination number = 4

(6) $Pyr_{48}Alm_{52}$

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$(Fe_{1.52}Mg_{1.41})\ Al_{1.99}Ca_{0.01}Si_{3.04}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.50955(6)Å, α=β=γ=90°
Volume (Å^3)	1524.67(2)
R_{int}	0.0391
All reflections	5808
Great reflections	115
Refined parameters	19
R_1	0.0175
wR_2	0.0505
S	1.0767

Atom	Occupancy	x	y	z	U_{iso}
Mg	0.451(18)	0.875	0.5	0.25	0.0076(4)
Fe	0.551(9)	0.875	0.5	0.25	0.0076(4)
Si	1	0.125	0.5	0.25	0.0037(5)
Al	1	0	0.5	0.5	0.0019(5)
O	1	0.03364(6)	0.54969(6)	0.34702(6)	0.0057(4)

[AlO_6] octahedron
Average bond length = 1.8913(7) Å
Octahedron volume = 9.0097 Å^3
Effective coordination number = 6
[FeO_8] /[MgO_8] dodecahedron
Average bond length = 2.2876(7) Å
bond I length = 2.2153(6) Å
bond II length = 2.3598(7) Å
Polyhedral volume = 20.5820 Å^3
Effective coordination number =7.6858
[SiO_4] tetrahedron
Average bond length = 1.6370(7) Å
Polyhedral volume = 2.2068 Å^3
Effective coordination number = 4

(7) $Pyr_{57}Alm_{43}$

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$(Fe_{1.36}Mg_{1.82})\ Al_{1.93}Ca_{0.01}Ti_{0.01}Si_{2.95}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.5013(4), α=β=γ=90°
Volume (Å^3)	1521.39(16)
R_{int}	0.0336

All reflections			5802		
Great reflections			114		
Refined parameters			19		
R_1			0.0204		
wR_2			0.0544		
S			1.0958		
Atom	Occupancy	x	y	z	U_{iso}
Mg	0.52(2)	0.875	0.5	0.25	0.0095(5)
Fe	0.484(12)	0.875	0.5	0.25	0.0095(5)
Si	1	0.125	0.5	0.25	0.0058(5)
Al	1	0	0.5	0.5	0.0042(5)
O	1	0.03365(8)	0.54984(8)	0.34696(8)	0.0082(4)

$[AlO_6]$ octahedron
Average bond length = 1.8912(10) Å
Octahedron volume = 9.0068 $Å^3$
Effective coordination number = 6
$[FeO_8]$ /$[MgO_8]$ dodecahedron
Average bond length = 2.2853(9) Å
bond I length = 2.2140(9) Å
bond II length = 2.3599(9) Å
Polyhedral volume = 20.514 $Å^3$
Effective coordination number =7.6937
$[SiO_4]$ tetrahedron
Average bond length = 1.6359(10) Å
Polyhedral volume = 2.2024 $Å^3$
Effective coordination number = 4

(8) **$Pyr_{67}Alm_{32}$**

Wavelength (Å)			0.4340		
Pressure (GPa)			0.0001		
Temperature (K)			298		
Composition			$(Fe_{1.03}Mg_{2.13})\ Al_{1.92}Ca_{0.01}Ti_{0.03}Si_{2.95}O_{12}$		
Symmetry			cubic, $Ia\bar{3}d$		
Lattice parameters			a=b=c=11.4887(3)Å, α=β=γ=90°		
Volume ($Å^3$)			1516.40(12)		
R_{int}			0.0411		
All reflections			5824		
Great reflections			120		
Refined parameters			19		
R_1			0.0213		
wR_2			0.0523		
S			1.1687		
Atom	Occupancy	x	y	z	U_{iso}
Mg	0.666(19)	0.875	0.5	0.25	0.0075(4)
Fe	0.334(9)	0.875	0.5	0.25	0.0075(4)
Si	1	0.125	0.5	0.25	0.0033(4)
Al	1	0	0.5	0.5	0.0025(4)
O	1	0.03342(7)	0.54989(7)	0.34683(6)	0.0050(4)

$[AlO_6]$ octahedron
Average bond length = 1.8901(8)Å
Octahedron volume = 8.9903 $Å^3$
Effective coordination number = 6
$[FeO_8]$ /$[MgO_8]$ dodecahedron
Average bond length = 2.2810(8) Å

bond I length = 2.2088(8) Å
bond II length = 2.3532(8) Å
Polyhedral volume = 20.4075 Å^3
Effective coordination number =7.684
[SiO_4] tetrahedron
Average bond length = 1.6349(8) Å
Polyhedral volume = 2.1970 Å^3
Effective coordination number = 4

(9) $Pyr_{78}Alm_{22}$

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$(Fe_{0.71}Mg_{2.49})\ Al_{1.85}Ca_{0.01}Si_{3.01}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c=11.48023(8)Å, α=β=γ=90°
Volume (Å^3)	1513.04(3)
R_{int}	0.0368
All reflections	5900
Great reflections	116
Refined parameters	19
R_1	0.0181
wR_2	0.0504
S	1.1291

Atom	Occupancy	x	y	z	U_{iso}
Mg	0.753(19)	0.875	0.5	0.25	0.0087(4)
Fe	0.244(9)	0.875	0.5	0.25	0.0087(4)
Si	1	0.125	0.5	0.25	0.0038(4)
Al	1	0	0.5	0.5	0.0036(4)
O	1	0.03331(6)	0.55001(6)	0.34670(6)	0.0058(4)

[AlO_6] octahedron
Average bond length = 1.8903(7) Å
Octahedron volume = 8.9914 Å^3
Effective coordination number = 6
[FeO_8] /[MgO_8] dodecahedron
Average bond length = 2.2779(7) Å
bond I length = 2.2057(7) Å
bond II length = 2.3501(7) Å
Polyhedral volume = 20.3269 Å^3
Effective coordination number =7.6832
[SiO_4] tetrahedron
Average bond length = 1.6340(7) Å
Polyhedral volume = 2.1923 Å^3
Effective coordination number = 4

(10) Pyr_{100}

Wavelength (Å)	0.4340
Pressure (GPa)	0.0001
Temperature (K)	298
Composition	$Mg_{3.12}Al_{1.96}Ti_{0.01}Si_{2.96}O_{12}$
Symmetry	cubic, $Ia\bar{3}d$
Lattice parameters	a=b=c= 11.45522(10)Å, α=β=γ=90°
Volume (Å^3)	1513.18(4)
R_{int}	0.0579

All reflections	6012
Great reflections	134
Refined parameters	17
R_1	0.0228
wR_2	0.0624
S	1.0938

Atom	Occupancy	x	y	z	U_{iso}
Mg	1	0.875	0.5	0.25	0.0086(4)
Si	1	0.125	0.5	0.25	0.0027(4)
Al	1	0	0.5	0.5	0.0032(4)
O	1	0.03298(7)	0.55039(6)	0.34677(6)	0.0045(4)

[AlO_6] octahedron
Average bond length = 1.8860(8) Å
Octahedron volume = 8.9264 Å^3
Effective coordination number =
[MgO_8] dodecahedron
Average bond length = 2.2779(8) Å
Average bond I length = 2.1993(8) Å
Average bond II length = 2.3400(7) Å
Polyhedral volume = 20.1007 Å^3
Effective coordination number =7.6980
[SiO_4] tetrahedron
Average bond length = 1.6350(8) Å
Polyhedral volume = 2.1954 Å^3
Effective coordination number = 4