Supplementary information
van Westrenen et al.

This contains (1) A comparison between observed and calculated structural parameters of the end members pyrope and grossular. (2) The GULP input file for configuration 1 (see text). All potential parameters are given in ref. 9. Radii for region 1 and 3 were 8 and 14 Å respectively.

(1) Comparison between observed and simulated lattice parameters of endmember garnets

| Property | Pyrope | | Grossular | | |
	Observed[a]	Simulated	Observed[b]	Simulated	(Unit)
Unit cell dimensions and oxygen atom coordinates					
a	11.452	11.281	11.848	11.874	(Å)
b	11.452	11.281	11.848	11.874	(Å)
c	11.452	11.281	11.848	11.874	(Å)
x(O)	0.0329	0.0318	0.0382	0.0385	-
y(O)	0.0503	0.0519	0.0453	0.0458	-
z(O)	0.6533	0.6519	0.6514	0.6493	-
Dodecahedron (X-site)					
X-O(1)	2.197	2.168	2.322	2.331	(Å)
X-O(2)	2.340	2.283	2.487	2.542	(Å)
<X-O>	2.269	2.225	2.405	2.437	(Å)
O4-O6	2.708	2.649	2.971	3.076	(Å)
O4-O7	2.778	2.721	2.859	2.923	(Å)
Octahedron (Y-site)					
Al-O	1.886	1.846	1.926	1.894	(Å)
O1-O4 shared	2.617	2.541	2.758	2.747	(Å)
O1-O5 unshared	2.716	2.678	2.689	2.613	(Å)
Tetrahedron (Z-site)					
Si-O	1.634	1.635	1.646	1.649	(Å)
O1-O2	2.497	2.505	2.572	2.580	(Å)
O1-O3	2.751	2.749	2.745	2.743	(Å)

[a] Lattice parameters for pyrope from Armbruster et al., Am. Min., 77, 512 (1992)
[b] Lattice parameters for grossular from Ganguly et al., Am. Min., 78, 583 (1993)

(2) GULP input file for configuration 1 (see text). Sizes of regions I and IIa were 10 Å and 18 Å respectively.

```
opti prop conp nosym full
cell
11.610 11.610 11.610 90. 90. 90. 1 1 1 0 0 0
frac
Mg   core   0.125000   0.000000   0.250000
Mg   core   0.625000   0.500000   0.750000
Mg   core   0.875000   0.500000   0.250000
Mg   core   0.375000   0.000000   0.750000
Mg   core   0.250000   0.125000   0.000000
Mg   core   0.750000   0.625000   0.500000
Mg   core   0.000000   0.250000   0.125000
Mg   core   0.500000   0.750000   0.625000
Mg   core   0.250000   0.875000   0.500000
Mg   core   0.750000   0.375000   0.000000
Mg   core   0.500000   0.250000   0.875000
Mg   core   0.000000   0.750000   0.375000
Ca   core   0.375000   0.500000   0.250000
Ca   core   0.875000   0.000000   0.750000
Ca   core   0.625000   0.000000   0.250000
Ca   core   0.125000   0.500000   0.750000
Ca   core   0.250000   0.375000   0.500000
Ca   core   0.750000   0.875000   0.000000
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Al   core   0.250000   0.250000   0.750000
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Si   core   0.875000   0.500000   0.750000
Si   core   0.125000   0.000000   0.750000
Si   core   0.625000   0.500000   0.250000
Si   core   0.250000   0.375000   0.000000
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Si   core   0.250000   0.625000   0.500000
Si   core   0.000000   0.750000   0.125000
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Si   core   0.750000   0.375000   0.500000
Si   core   0.250000   0.875000   0.000000
Si   core   0.250000   0.125000   0.500000
Si   core   0.750000   0.625000   0.000000
Si   core   0.375000   0.500000   0.750000
Si   core   0.875000   0.000000   0.250000
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Si   core   0.125000   0.500000   0.250000
Si   core   0.625000   0.000000   0.750000
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Si   core   0.000000   0.750000   0.625000
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(2) Continued

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O	core	0.051913	0.651869	0.031827
O	core	0.551913	0.151869	0.531827
O	core	0.348131	0.468173	0.051913
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O	core	0.151869	0.468173	0.948087
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O	core	0.348131	0.531827	0.448087
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O	core	0.718173	0.098131	0.698087
O	core	0.598131	0.198087	0.218173
O	core	0.098131	0.698087	0.718173
O	core	0.781827	0.901869	0.198087
O	core	0.281827	0.401869	0.698087
O	core	0.598131	0.801913	0.281827
O	core	0.098131	0.301913	0.781827
O	core	0.281827	0.598131	0.801913
O	core	0.781827	0.098131	0.301913
O	core	0.401869	0.301913	0.218173
O	core	0.901869	0.801913	0.718173
O	core	0.218173	0.401869	0.301913
O	core	0.718173	0.901869	0.801913
O	core	0.901869	0.198087	0.781827
O	core	0.401869	0.698087	0.281827
O	core	0.468173	0.448087	0.848131
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O	core	0.031827	0.551913	0.348131
O	core	0.531827	0.051913	0.848131
O	core	0.968173	0.051913	0.151869
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O	core	0.031827	0.448087	0.151869
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O	core	0.848131	0.468173	0.448087
O	core	0.348131	0.968173	0.948087
O	core	0.448087	0.848131	0.468173
O	core	0.948087	0.348131	0.968173
O	core	0.151869	0.031827	0.448087
O	core	0.651869	0.531827	0.948087
O	core	0.551913	0.348131	0.031827
O	core	0.051913	0.848131	0.531827
O	core	0.348131	0.031827	0.551913
O	core	0.848131	0.531827	0.051913
O	core	0.051913	0.151869	0.968173
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O	core	0.151869	0.968173	0.051913

(2) Continued

O	core	0.651869	0.468173	0.551913
O	core	0.448087	0.151869	0.031827
O	core	0.948087	0.651869	0.531827
O	core	0.301913	0.281827	0.901869
O	core	0.801913	0.781827	0.401869
O	core	0.698087	0.218173	0.901869
O	core	0.198087	0.718173	0.401869
O	core	0.198087	0.281827	0.098131
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O	core	0.781827	0.401869	0.801913
O	core	0.901869	0.301913	0.281827
O	core	0.401869	0.801913	0.781827
O	core	0.718173	0.598131	0.301913
O	core	0.218173	0.098131	0.801913
O	core	0.901869	0.698087	0.218173
O	core	0.401869	0.198087	0.718173
O	core	0.218173	0.901869	0.698087
O	core	0.718173	0.401869	0.198087
O	core	0.098131	0.198087	0.281827
O	core	0.598131	0.698087	0.781827
O	core	0.281827	0.098131	0.198087
O	core	0.781827	0.598131	0.698087
O	core	0.598131	0.301913	0.718173
O	core	0.098131	0.801913	0.218173
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O	shel	0.537262	0.546039	0.156156
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O	shel	0.962738	0.453961	0.656156
O	shel	0.537262	0.453961	0.343844
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O	shel	0.462738	0.046039	0.343844
O	shel	0.962738	0.546039	0.843844
O	shel	0.656156	0.037262	0.046039
O	shel	0.156156	0.537262	0.546039
O	shel	0.046039	0.656156	0.037262
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O	shel	0.343844	0.462738	0.046039
O	shel	0.843844	0.962738	0.546039
O	shel	0.953961	0.156156	0.462738
O	shel	0.453961	0.656156	0.962738
O	shel	0.156156	0.462738	0.953961
O	shel	0.656156	0.962738	0.453961
O	shel	0.453961	0.343844	0.537262
O	shel	0.953961	0.843844	0.037262
O	shel	0.343844	0.537262	0.453961
O	shel	0.843844	0.037262	0.953961
O	shel	0.046039	0.343844	0.462738
O	shel	0.546039	0.843844	0.962738
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O	shel	0.796039	0.287262	0.593844
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O	shel	0.796039	0.712738	0.906156
O	shel	0.203961	0.787262	0.906156
O	shel	0.703961	0.287262	0.406156
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O	shel	0.712738	0.093844	0.703961
O	shel	0.593844	0.203961	0.212738
O	shel	0.093844	0.703961	0.712738
O	shel	0.787262	0.906156	0.203961
O	shel	0.287262	0.406156	0.703961
O	shel	0.593844	0.796039	0.287262
O	shel	0.093844	0.296039	0.787262
O	shel	0.287262	0.593844	0.796039
O	shel	0.787262	0.093844	0.296039

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O    shel    0.712738    0.906156    0.796039
O    shel    0.906156    0.203961    0.787262
O    shel    0.406156    0.703961    0.287262
O    shel    0.462738    0.453961    0.843844
O    shel    0.962738    0.953961    0.343844
O    shel    0.037262    0.546039    0.343844
O    shel    0.537262    0.046039    0.843844
O    shel    0.962738    0.046039    0.156156
O    shel    0.462738    0.546039    0.656156
O    shel    0.037262    0.453961    0.156156
O    shel    0.537262    0.953961    0.656156
O    shel    0.843844    0.462738    0.453961
O    shel    0.343844    0.962738    0.953961
O    shel    0.453961    0.843844    0.462738
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O    shel    0.656156    0.537262    0.953961
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O    shel    0.546039    0.656156    0.462738
O    shel    0.156156    0.962738    0.046039
O    shel    0.656156    0.462738    0.546039
O    shel    0.453961    0.156156    0.037262
O    shel    0.953961    0.656156    0.537262
O    shel    0.296039    0.287262    0.906156
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O    shel    0.703961    0.787262    0.593844
O    shel    0.296039    0.712738    0.593844
O    shel    0.796039    0.212738    0.093844
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O    shel    0.406156    0.796039    0.787262
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O    shel    0.906156    0.703961    0.212738
O    shel    0.406156    0.203961    0.712738
O    shel    0.212738    0.906156    0.703961
O    shel    0.712738    0.406156    0.203961
O    shel    0.093844    0.203961    0.287262
O    shel    0.593844    0.703961    0.787262
O    shel    0.287262    0.093844    0.203961
O    shel    0.787262    0.593844    0.703961
O    shel    0.593844    0.296039    0.712738
O    shel    0.093844    0.796039    0.212738
space
1
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Mg    core    2.000000
Si    core    4.000000
Al    core    3.000000
O     core    0.869020
O     shel   -2.869020
buck
Ca    core O    shel 1090.4000    0.343700 .00000E+00 0.000 12.000
buck
Mg    core O    shel 1428.5000    0.294500 .00000E+00 0.000 12.000
buck
Al    core O    shel 1114.9000    0.311800 .00000E+00 0.000 12.000
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(2) Continued

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buck
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spring
O   74.920000
three
Si   core O   shel O   shel 2.0930    109.49000  1.900  1.900  3.000
dump forfull
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