

A Calorimetric Study of the $\text{Mg}_3(\text{Mg},\text{Si})\text{Si}_3\text{O}_{12}$ (majorite)- $\text{Mg}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ (pyrope) Garnet Solid Solution

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Garnet is a major constituent of the upper mantle, transition zone and the top region of the lower mantle. Over this range of stability, spanning from 2-28 GPa, garnet compositions change significantly as a result of reactions between other phases of the mantle. At shallow upper mantle conditions, garnet is a solid solution with major element substitutions that are described by the formula $(\text{Mg},\text{Fe},\text{Ca})_3\text{Al}_2\text{Si}_3\text{O}_{12}$. Above approximately 4 GPa substitution of Mg, Fe and Si onto the octahedrally-coordinated Al site occurs causing MgSiO_3 and FeSiO_3 pyroxene components to dissolve into the garnet structure as majorite components and these transitions are described by the garnet systems $\text{Mg}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ - $\text{Mg}_4\text{Si}_4\text{O}_{12}$ and $\text{Fe}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ - $\text{Fe}_4\text{Si}_4\text{O}_{12}$. As silicate perovskites are formed in the mantle by reactions that involve the breakdown of majoritic garnet, thermodynamic parameters along this garnet solid solution are essential for modeling these reactions.

We have measured the enthalpy of dissolution of garnet solid solution in the system $\text{Mg}_3(\text{Mg},\text{Si})\text{Si}_3\text{O}_{12}$ (majorite)- $\text{Mg}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ (pyrope) by lead borate oxide drop solution calorimetry in order to independently assess the non ideality of the garnet solid solution and to estimate the thermodynamic properties of the fictive $\text{Mg}_3(\text{Mg},\text{Si})\text{Si}_3\text{O}_{12}$ majorite end member. Measured values of enthalpies of dissolution show that the enthalpy of dissolution decreases non-linearly with increasing majorite content starting from pyrope end member. A sharp break in the trend was recorded for the enthalpies of dissolution between pyrope contents of 32 mol% and 24 mol% close to the symmetry-breaking cubic-tetragonal transition in garnets. The data imply that $\text{Mg}_3(\text{Mg},\text{Si})\text{Si}_3\text{O}_{12}$ majorite- $\text{Mg}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ pyrope solid solution has strong non-ideal mixing properties. Thermodynamic treatment of the data yields a value -37 kJ/mol as the enthalpy of solution of the fictive cubic majorite garnet.