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Volumes of mixing in aluminosilicate garnets: Solid solution and strain behavior

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ABSTRACT

The volumes of mixing of six garnet binaries in the four-component system almandine-pyrope-spessartine-grossular have been analyzed and compared. The almandine-pyrope join is thermodynamically ideal and the others show positive deviations from ideality. The magnitude of the excess volume of mixing of a given binary, as described by a symmetric mixing model, is shown to be a function of the volume difference between the two end-member components. The relationship between the magnitude of nonideality and volume difference can be described with a linear or quadratic function, but the latter has a physical basis in strain theory. Positive deviations from ideal thermodynamic mixing are primarily a result of strain resulting from the substitution of dodecahedral X-site cations of different sizes, but second-order chemical effects (e.g., electronic, coulombic) also appear to contribute to thermodynamic properties. XRD and IR spectroscopic measurements show that nonlinear behavior of microscopic properties, such as bond lengths and polyhedral distortions and volumes, across a solid solution is strong for pyrope-grossular garnets and very small to nonexistent for almandine-pyrope solid solutions. The magnitude of the deviations from linearity in the microscopic properties can be related to the excess volume of mixing for a given binary. A good correlation exists between microscopic structural strain and the macroscopic volume of mixing.