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Speciation of P in and partitioning between aqueous fluids and silicate melts to upper mantle temperatures and pressures Speciation of P in and partitioning between aqueous fluids and silicate melts to upper mantle temperatures and pressures

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Evidence from high-grade metamorphic terranes suggest that phosphorus solubility and solution mechanisms in silicate melts and aqueous fluids depend on composition resulting from a range of possible P-bearing structural complexes. The structure of phosphorus-bearing, H₂O-saturated silicate melts, silicate-saturated aqueous fluids, and silicate-rich single phase (supercritical) liquids has been characterized, therefore, via in-situ experiments from ambient temperatures and pressures to 800°C/1486 MPa. The solution mechanisms and partitioning of structural species between fluids and melts were determined with the aid confocal microRaman and with FTIR spectroscopy backed up with published phosphorus-31 MAS NMR data [1]. The experiments were conducted in an Ir-gasketed hydrothermal diamond anvil cell. Temperature and pressure were recorded with thermocouples (1°C uncertainty) and pressure- and temperature-dependent Raman shift of ¹³C diamonds (40 MPa uncertainty). Starting materials were aluminum-free Na₂O·4SiO₂ (NS4) and with 10 mol % Al₂O₃ (NA10) substituting for SiO₂, both with 5 mol % P₂O₅. These compositions enabled characterization of phosphorus behavior with coexisting haploandesite melt and aluminosilicate-saturated aqueous fluids with variable Al-content.

Aluminosilicate species of Q₀, Q₁, Q₂, and Q₃ type exist in coexisting fluid and melt and in single phase liquid together with phosphate species, PO₄, P₂O₇, and Q_nP. In the Q_nP notation, the n-value denotes the number of oxygen in the structural species shared with P and Si. Al substitutes for Si predominantly in the Q_nP species. In melts, the abundance of the most depolymerized silicate species, Q₀, is positively correlated with temperature and pressure, whereas that of the most polymerized species, Q₃, decreases with temperature and pressure. In the silicate solute in aqueous fluids, the opposite relationship exists with Q₀ abundance decreasing and Q₃ (and Q₁ and Q₂) abundance increasing with increasing temperature and pressure. The silicate melts, therefore, become increasingly depolymerized and the silicate solute in aqueous fluids decreasingly depolymerized. The P₂O₇ and Q_nP are the dominant phosphate species in fluid, melt, and single phase liquid with orthophosphate, PO₄, playing a subordinate role. The fluid/melt partition coefficients for P₂O₇ and Q_nP species are in the 0.15-0.7 range with that of Q_nP being greater than that of P₂O₇. The PO₄ fluid/melt partition coefficients are <0.2. In all cases, the partition coefficients increase with increasing temperature and pressure. There is no clear influence of Al₂O₃. Hence, it appears that P-bearing complexes in fluids and melts are associated with Na⁺. Mobility of phosphorus during metamorphic processes likely is principally governed by availability of alkali metals (and perhaps alkaline earths), whereas Al/Si-ratio may be a less important composition parameter. The P-partitioning between fluids and melts likely are significantly pressure-dependent because fluid and melt speciation is sensitive to pressure.

[1] Cody, B. O., Mysen, B. O., Saggi-Szabo, G., and Tossell, J. A., 2001. *Geochim. Cosmochim. Acta* 65, 2395.

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