

Characterisation of quenched carbonate-silicate melts by vibrational spectroscopy, XAFS and HEXRD: Structure and Y, Sr, Zr speciation

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Carbonatite and carbonate-silicate melts show very distinct physical properties compared to silicate melts, i.e. lower density and viscosity and very high electrical conductivity. These properties are a consequence of differences in their structural properties. There are two poorly understood key aspects of the evolution of carbonate-silicate systems: (1) transitions between carbonate-rich and silicate-rich varieties with increasing temperature during partial melting and (2) the possibility of liquid immiscibility between carbonate and silicate melts.

Melting experiments at 1500°C/5.0 GPa and 1700°C/5.5GPa were performed on mixtures CaCO₃ + CaSiO₃ with 10, 20, 30 and 50 wt.% of the carbonate component to trace the structural changes accompanying variable carbonate/silicate ratio in melts. Minor SrCO₃, Y₂O₃, ZrO₂ and Nb₂O₅ were added to investigate local environment of the trace elements in the quenched melts. The quenched samples were characterized by electron microprobe (EMP), IR and Raman microscopy, EXAFS at Sr, Y, Zr K-edges and pair distribution function (PDF) analysis.

EMP mapping of the quenched samples shows homogeneous distribution of Sr and Y on the micron scale. IR and Raman reveal presence of silicate glass and disordered and fine-crystalline carbonates. The ν_1 (CO₃)²⁻ band is downshifted from pure calcite, indicating distorted environment. XAFS shows a complete decomposition of SrCO₃, ZrO₂ and Y₂O₃ in the melts. Higher PT conditions modify the Sr local coordination from “carbonate-like” to more “silicate-like”. The quench kinetics, however, may also play a role. The ratio CaSiO₃/CaCO₃ does not affect the Sr coordination, but likely modifies the Y environment. Carbonate-rich compositions favor cubic coordination of Y in the melts. The Zr environment closely resembles Zr in silicate glass.