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Temperature Dependence of Iron-Redox Equilibria in Ultra-High Temperature Melts

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The Fe^{3+}/Fe^{2+} ratio is critical for magmatic redox evolution and depends on temperature. This dependence is particularly important for magma oceans, where temperatures (2500-4000 K) far exceed previous experimental constraints. To date, thermodynamic models of magmatic Fe^{3+}/Fe^{2+} are calibrated using experiments restricted to the narrow superliquidus range (1200–1600 °C), where the log (Fe^{3+}/Fe^{2+})–$1/T$ relationship is approximately linear. Here we extend investigations to ultra-high temperatures using aerodynamic laser levitation experiments in an O_2 atmosphere, enabling quenching of synthetic komatiitic ($FeO–SiO_2–CaO–Al_2O_3–MgO$) and picritic ($FeO–SiO_2–CaO–Al_2O_3–MgO–TiO_2$) melts from 1350 °C to 2600 °C at fixed oxygen fugacity (f_{O2}).Both oxidized and reduced starting materials were used and a time series was conducted to monitor approaches to equilibrium.

Quenched glasses were analyzed by EPMA and Mössbauer spectroscopy, revealing dominant paramagnetic signals from the glass and minor ferromagnetic or superparamagnetic components derived from oxide quench phases. Vibrating Sample magnetometry and MPMS-XL magnetometry allowed identification of these nanocrystalline oxides. The combined results show that at log f_{O2}=0, Fe^{3+}/Fe^{T} decreases from 0.81 at 1350 °C to 0.57 at 2600 °C for picrite, and from 0.71 at 1460 °C to 0.43 at 2600 °C for komatiite. The log (Fe^{3+}/Fe^{2+})–$1/T$ relationship becomes non-linear, particularly above 2100 °C, consistent with the Hirschmann (2022) thermodynamic model which incorporates differences in heat capacity (ΔC_P) between FeO and $FeO_{1.5}$ in melt. At high temperatures, melts are richer in Fe^{3+} than previously predicted. New temperature-dependent parameterizations are derived for both compositions. The offset between picrite and komatiite trends demonstrates a systematic compositional control on redox equilibria.

Extrapolation to metal-saturated conditions ($\Delta IW = -2$) indicates that Fe^{3+}/Fe^{T} increases with temperature more strongly than predicted by existing models. Using EOS parameters for 12.5 mol% Fe in $(Mg,Fe)SiO_3$ from Deng et al. (2020) to account for pressure effects, the new parameterization predicts that along a magma-ocean geotherm (T_P = 2500 K), Fe^{3+}/Fe^{T} increases from 0.08 to 0.12 for komatiite (0.13–0.27 for picrite) between 28 and 53 GPa. In an isochemical convecting magma ocean, these correspond to surface f_{O2}s of IW–0.8 to IW for (IW–0.5 to IW+1.7 for picrite).