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Fractional crystallization by itself cannot explain the elevated oxidation state of arc magmas

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Arc magmas are globally oxidized, but whether this results from an oxidized mantle source or from fractional crystallization remains debated [1,2]. Resolving this requires assessing whether crystallization can oxidize magmas across the full range of arc pressures. This critically depends on the Fe^{3+}/Fe^{2+} exchange coefficients (Kd) of the dominant crystallizing phases, particularly amphibole and clinopyroxene at mid-to-low pressures, for which data are poorly constrained.

We performed piston–cylinder experiments at 1.25 GPa and 900–1000 °C with oxygen fugacity buffered ranging from CCO to Ru-RuO_2. The $Fe^{3+}/\Sigma Fe$ ratios of the quenched silicate melt and coexisting minerals, including amphibole, clinopyroxene, and orthopyroxene, were quantified by synchrotron Mössbauer spectroscopy collected at beamline ID14 at ESRF and fitted with the SyncMoss software package[3]. The resolved Kd of 1.8–3.7 for amphibole and 1.1–2.5 for clinopyroxene, 0.3-0.7 for orthopyroxene. Integrating these new data with existing garnet values (Kd < 1)[2], we modeled fractional crystallization of a primary arc basalt from 0.5 to 2.0 GPa. Redox evolution is controlled by pressure via the crystallizing assemblage: at low pressures (0.5–1 GPa), amphibole ± clinopyroxene crystallization does not increase melt oxygen fugacity, whereas at high pressures (1.25–2 GPa), clinopyroxene + garnet leads to only a slight increase (<0.2 log units). Critically, the high Kd of clinopyroxene counteracts any significant oxidation even when garnet is present.

Therefore, fractional crystallization does not universally oxidize arc magmas. The pressure-dependent mineral assemblage, now constrained by key amphibole and clinopyroxene data, precludes crystallization as a global oxidation mechanism, providing strong support for inheritance from an oxidized mantle source.

[1] Tang et al. (2018) Sci. Adv. 4, eaar4444.

[2] Holycross & Cottrell (2023) Science 380, 506-509.

[3] Yaroslavtsev et al. (2023) J. Synchrotron Rad. 30, 596-604.