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Clinopyroxene records of magma redox: insights from experiments and thermodynamic modelling

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Iron plays a central role in both setting and recording magma redox states, which are commonly expressed in terms of oxygen fugacity deviations from known equilibria such as fayalite-magnetite-quartz (FMQ). Magmas erupted in different tectonic settings are characterised by different redox states, which have implications for understanding how magmatic evolution, ore mineralisation and volcanic degassing vary in space and time. For example, mid-ocean ridge basalts are relatively reduced (FMQ), arc basalts relatively oxidised (up to FMQ+2) and ocean island basalts are often but far from universally oxidised (<FMQ to ≥FMQ+2). However, current tools for estimating magma redox conditions (e.g., Fe-XANES on glasses, two-oxide oxybarometry) are subject to diverse limitations, making it challenging to interrogate the nature and origins of redox variability between different magmatic systems. Fortunately, recent studies have demonstrated that electron probe microanalysis can return robust estimates of clinopyroxenes Fe^{2+} and Fe^{3+} contents from stoichiometry when the analyses are performed with sufficient care. Importantly, this improved ability to measure clinopyroxene crystals offers new ways of investigating variability in Fe valence, and thus redox, within and between natural and experimental magmas. Unfortunately, relating the valence state of Fe to magma redox is challenging because of steric effects imposed by the clinopyroxene structure. In striving to overcome these challenges, we present a combination of experimental observations and thermodynamic calculations designed to investigate the incorporation of Fe into clinopyroxene crystals at magmatic conditions. Experiments and calculations were performed on Icelandic and Azorean basalts and tephrites under a range of redox conditions (ca. FMQ-2 to FMQ+2) at magmatic temperatures (ca. 1150 °C) and pressures (100–500 MPa) using internally heated pressure vessels (IHPVs) and the MAGEMin Gibbs energy minimising software with the newest generation of thermodynamic models. Both experiments and calculated phase diagrams demonstrate that clinopyroxene Fe^{3+} contents are sensitive to prevailing redox conditions and can thus preserve information about magmatic oxygen fugacity in the geological record. Nevertheless, more work is required to disentangle the effects of pressure, oxygen fugacity, magma composition and kinetics on Fe valance in magmatic clinopyroxene crystals.