Redox and Magma Ocean Evolution:Insights from Experimental Studies

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The redox state of the early Earth's magma ocean (MO) was pivotal in controlling the speciation and solubility of volatile elements (C,N,S,H), affecting their degassing to the primitive atmosphere and, in turn, MO crystallization and subsequent geodynamic evolution. This redox evolution also significantly influenced the composition of the Hadean atmosphere.

 Fe_2O_3 produced in a deep MO in equilibrium with coredestined alloy sets the early redox budget. Previous experiments (\leq 28 GPa)^{1,2} and first-principles calculations³ indicated that a deep terrestrial MO produces appreciable Fe^{3+} , but predict Fe^{3+}/Fe^T that conflict by an order of magnitude. We measured Fe^{3+}/Fe^T of glasses quenched from melts equilibrated with Fealloy with laser heated diamond anvil cell (LHDAC) at 38-71 GPa, 3600-4400K, analyzed by synchrotron Mössbauer spectroscopy (SMS)⁴. These indicate Fe^{3+}/Fe^T of 0.056-0.112 in a terrestrial magma ocean with mean alloy-silicate equilibration pressures of 28-53 GPa, producing sufficient Fe_2O_3 to account for the modern bulk silicate Earth redox budget and surficial conditions near or more oxidizing than the iron-wüstite buffer, which would stabilize a primitive CO- and H_2O - rich atmosphere.

Bridgmanite, the lower mantle majority phase, incorporates appreciable Fe³⁺, even under reducing conditions, influencing the redox state of the lower mantle and affects the oxidation of the residual MO and overlying atmosphere as they evolve^{5,6}. The exchange coefficients of Fe³⁺ and Fe²⁺ between Brg and melts are the crucial parameters for evaluating redox mass transfer during MO crystallization. To investigate this partitioning, we conducted LHDAC experiments that produced coexisting Brg and melt, and analyzed both phases with the new ~1 mm SMS beam at ESRF. The resolved exchange coefficients range from 1.66 to 5.07, which is broadly consistent with previous Brg/melt K_D measurements from previous multi anvil experiments^{5,6}, indicating during deep crystallization, MO will remain alloy-saturated until low pressure phases replace bridgmanite on the liquidus.

Reference:

[1] Armstong et al. (2019) Science 365, 903-906; [2] Kuwahara et al. (2023) Nat. Geosci. 16, 461-465; [3] Deng et al. (2020) Nat. Commun. 11:2007; [4] Zhang et al. (2024) Sci. Adv. 10, eadp1752; [5] Huang et al. (2023) ESPL 624, 118447;[6] Kuwahara and Nakada (2023) ESPL 615,118197