

GALLIUM ISOTOPIC HETEROGENEITY IN LUNAR HIGHLAND ROCKS

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Introduction: The Moon is highly depleted in volatile elements compared to both chondritic meteorites and the Earth, and this is often attributed to a global-scale evaporation event [*e.g.*, 1-2]. One way to provide key insights into the nature of this devolatilization is by investigating mass-dependent isotope variations of moderately volatile elements (MVEs). During evaporative loss, lighter isotopes preferentially enter the vapor phase, leaving behind a residue that is relatively enriched in heavy isotopes. One of these MVEs is gallium (Ga), which is lithophile, has two stable isotopes, and a 50% condensation temperature of 968K [3]. Previous work has shown that lunar mare basalts are indeed isotopically heavy ($\delta^{71}\text{Ga} = 0.33 \pm 0.16\%$, [4-5], where $\delta^{71}\text{Ga}$ is the deviation from terrestrial Ga isotope ratios in permil) compared to the bulk silicate Earth ($\delta^{71}\text{Ga} = 0.00 \pm 0.06\%$, [6]). In contrast, the limited number of lunar highland rocks that have been analyzed to this day appear to be far more variable in their Ga isotopic signatures. Considering that highland rocks make up ~80% of the lunar crust, these samples are crucial to understand the lunar volatile depletion. In this work, we report Ga isotopic as well as major and trace element data for a suite of lunar highland rocks, to further explore the circumstances under which MVE loss could have taken place.

Samples and Methods: The sample set investigated here includes six rocks from the ferroan anorthosite suite (FAS) and four magnesian suite (MGS) rocks, and is interpreted together with a previously published dataset for lunar highland rocks [5]. In addition, we investigate an impact-derived melt glass that originally coated a host FAS rock. Given that this impact melt contains material that was volatilized and redistributed during one or multiple impact(s), it is expected to contain isotopically light Ga. Chemical purification and isotopic measurements of Ga were performed following previously established methods [7] and using the ThermoScientific Neptune Plus MC-ICPMS at LLNL. Prior to chemical separation, a 5% aliquot was taken per sample for major and trace element analyses using a ThermoScientific Element Plus XR-ICPMS at LLNL.

Results: The concentrations of many elements in the FAS and MGS are correlated with one another, consistent with both lunar rock suites being comprised of variable mixtures of multiple chemically distinct components (*e.g.*, plagioclase, mafic minerals, and KREEP). Comparable to previous studies [4-5], we find that MGS rocks are isotopically similar to mare basalts in Ga, whereas FAS display a range of Ga isotopic compositions that is both lighter and heavier compared to the mare basalts, MGS and the BSE. The impact melt shows the heaviest Ga isotopic composition in our sample set and its chemical composition resembles KREEP-like material, similar to other previously described impact melts [9-10].

Discussion: Given that the intrusive MGS and the extrusive mare basalts are assumed to derive from analogous lunar magma ocean (LMO) cumulates and have indistinguishable Ga isotope systematics, this argues against volatile degassing during eruption of the mare basalts as the primary cause for their relatively heavy Ga isotopic compositions. Similarly, the substantial Ga isotope variability of FAS—including both heavier and lighter isotopic compositions relative to the Earth—argues against global-scale evaporative loss of Ga from the Moon, *e.g.*, during lunar formation or LMO degassing [12-14]. Finally, the heavy Ga isotopic composition of the impact melt investigated here is opposite of what would be expected for locally devolatilized and secondarily redistributed Ga and difficult to reconcile with impact metamorphism being responsible for the observed Ga isotope variability, as it has been previously suggested for other MVEs (*e.g.*, Zn [11]). Instead, this composition may be related to admixture of KREEP-like material with an intrinsically heavy Ga isotopic signature.

Collectively, and when combining the Ga isotope data with trace element abundances, the observed Ga isotope variability in the FAS is best explained through igneous processes during progressive crystallization of the LMO. Given that the bulk of the Ga inventory in FAS (and MGS) is hosted in plagioclase [*e.g.*, 8], a viable explanation for the Ga isotope heterogeneity of the Moon is therefore the preferential incorporation of light Ga isotopes in the plagioclase crystal structure. Once this isotope fractionation from crystallization and extraction of anorthite during LMO solidification is accounted for, the Ga isotopic compositions of the Moon and the Earth overlap within uncertainty. The depletion of Ga (and perhaps other volatiles) from the Moon could, therefore, simply be inherited from its progenitor protoplanet(s), similar to considerations from [15].

References: [1] Canup and Asphaug (2001), *Nature* [2] Day and Moynier (2014) *Philos. Trans. Royal Soc.* [3] Lodders (2003), *ApJ* [4] Kato and Moynier (2017), *Sci. Adv.* [5] Wimpenny et al., (2022) *EPSL* [6] Kato et al., (2017) *Chem. Geol.* [7] Wimpenny et al. (2020) *GCA* [8] Sio et al., (2020) *EPSL* [9] Warren (1988) *LPSC XVIII* [10] McIntosh et al. (2020) *EPSL* [11] Kato et al. (2015) *Nature Comms.* [12] Sossi et al. (2018) *PNAS*. [13] Day and Moynier (2014) *Philos. Trans. Royal Soc.* [14] Tartèse et al. (2021) *PNAS*. [15] Borg et al. (2022) *PNAS*.