IMPLICATIONS FOR THE ORIGIN OF THE EARTH-MOON SYSTEM FROM GALLIUM ISOTOPES.

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Introduction: The Moon is highly depleted in volatile elements compared to both chondritic meteorites and the Earth, however, the timing of volatile loss is still debated [*e.g.,* 1-2]. One way to further elucidate the nature of this devolatilization is by investigating mass-dependent isotope variations of moderately volatile elements (MVE). During evaporative loss, lighter isotopes preferentially enter the vapor phase, leaving behind a residue that is relatively enriched in heavy isotopes. One of these MVE is gallium (Ga), which is lithophile, has two stable isotopes, and a 50% condensation temperature of 1010K [3]. Previous work has shown that Ga in lunar mare basalts is isotopically heavier (δ^{71}Ga = 0.33 ± 0.16‰, [4-5], where δ^{71}Ga is the deviation from terrestrial Ga isotope ratios in permil) than Ga in the bulk silicate Earth (δ^{71}Ga = 0.00 ± 0.06‰, [6]). However, correlations between δ^{71}Ga and trace element abundances indicate that magmatic processes have affected the Ga isotopic ratios of the mare basalt suite, and recent work suggested that the Ga isotopic composition of the Moon may be more similar to the Earth than previously thought [5]. So far, only a limited number of Ga isotopic analyses have been made from lunar highland rocks, yielding Ga isotopic compositions that are far more variable than those in the mare basalt suite [4, 5]. Considering that the lunar highlands make up ~80% of the lunar crust and contain a large proportion of the lunar Ga inventory [5], these samples are crucial to understanding Ga isotope systematics in the Moon. Here we report Ga isotopic data from a suite of lunar highland rocks to further explore the circumstances under which Ga and MVE loss took place.

Samples and Methods: The samples investigated here include six rocks from the ferroan anorthosite suite (FAS) and four magnesian suite (MGS) rocks, which are interpreted together with previously published data for highland rocks [5]. In addition, we investigate an impact-derived melt glass that originally coated a host FAS rock. This impact melt contains material that was volatilized and redistributed during one or multiple impact(s) and, as such, may 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-ICP-MS at LLNL. Prior to chemical separation, a 5% aliquot was taken per sample to determine major and trace element abundances using a ThermoScientific Element XR ICP-MS at LLNL.

Results: The abundances of many elements in the FAS and MGS are correlated, consistent with the idea that both lithologies are mixtures of chemically distinct components (e.g., plagioclase, mafic minerals, and KREEP). Despite the high modal abundance of anorthite (>90%), there are disproportionate variations of some trace elements in the bulk FAS, indicating significant variability in the plagioclase trace element chemistry.

The Ga isotopic compositions obtained for BCR-2, BHVO-2, and AGV-2 are consistent with estimates for the bulk terrestrial δ^{71}Ga value from [6] and [7], allowing for comparisons between the datasets published hitherto. Analogous to previous observations [4-5], we find that rocks from the MGS are isotopically similar to mare basalts, whereas FAS rocks display a range of Ga isotopic compositions that is both lighter and heavier compared to the mare basalts, MGS and the BSE (Fig. 1). The impact melt shows the heaviest Ga isotopic composition in our sample set and chemically resembles KREEP-like materials, similar to other previously described impact melts [8].

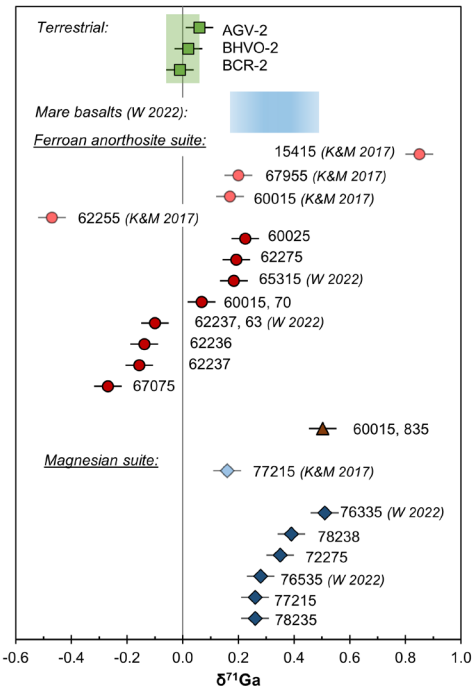

Figure 1: Gallium isotopic compositions of terrestrial and lunar samples investigated in this study. Sample 60015,835 (brown triangle) is an impact-derived melt glass. Gallium isotope data from [4] and [5] are shown for comparison.

Discussion: An average of the FAS investigated here is δ^{71}Ga = 0.00 ± 0.38 (2 s.d.), coinciding with the

terrestrial Ga isotopic composition [6], while showing considerably more variation. Given that the FAS contains most of the lunar Ga inventory, the substantial Ga isotope variability of FAS—straddling the Ga isotopic composition of the Earth—argues against global-scale evaporative loss of Ga from the Moon, e.g., during lunar formation or LMO degassing [9-11]. Similarly, considering that the MGS and the lunar mare basalts are assumed to derive from analogous lunar magma ocean (LMO) cumulates and display comparable δ^{71}Ga average values, this argues against degassing of Ga during eruption of the mare basalts as the primary cause for their relatively heavy δ^{71}Ga values. Lastly, the heavy Ga isotopic composition of the impact melt investigated here is opposite of the expected isotopic effect associated with degassing and condensation processes, which were inferred to explain the behavior of other MVEs, such as Zn [12]. Instead, its composition may be related to admixture of KREEP-like material with an intrinsically heavy Ga isotopic signature [5]. Collectively, and although Ga is defined as a moderately volatile element, these observations indicate that Ga isotope variations are not primarily controlled by devolatilization processes.

In contrast, both the Ga isotopic compositions and trace element abundances of the bulk FAS correspond with the An# of the plagioclase [13]. Given the importance of plagioclase during LMO crystallization [5] as well as the fact that most of the Ga in FAS (and MGS) is incorporated into plagioclase [*e.g.,* 14], another explanation for the observed Ga isotope heterogeneity is the preferential incorporation of light Ga isotopes in the plagioclase crystal structure during the later solidification stages of the LMO. This mechanism results in a relative enrichment of isotopically heavy Ga in the parental melt and is consistent with the previously observed correlation between δ^{71}Ga and the europium anomaly (Eu*) in the mare basalts [5]. Collectively, the currently available Ga isotope data therefore argue for igneous processes being mainly responsible for the observed lunar Ga isotope heterogeneity.

Using Ga isotope partitioning models, we find that $\Delta^{71}Ga_{\text{plagioclase-melt values}}$ of -0.30 to -0.40‰ can account for the range of Ga isotopic compositions in FAS and explain relatively high δ^{71}Ga values in the MGS, mare basalts and KREEP-rich samples. These models suggest a δ^{71}Ga value between 0.03 – 0.13‰ for the composition of the bulk Moon, which is potentially heavier, but within uncertainty of current estimates for the Ga isotopic composition of the Earth. Although the Moon is more depleted in MVEs than the Earth [e.g., 11], such similarity in their Ga isotopic compositions can be reconciled if their precursor bodies both formed in hot, volatile depleted regions of the inner Solar System. This would be supported by new constraints from Rb-Sr isotope systematics that indicate that the Moon's volatile deficit was inherited from Theia [16]. Finally, given the predicted gradient in nucleosynthetic isotope anomalies with heliocentric distance [17], this hypothesis is also consistent with the non-mass-dependent isotope overlap between the Moon and the Earth [e.g., 18].

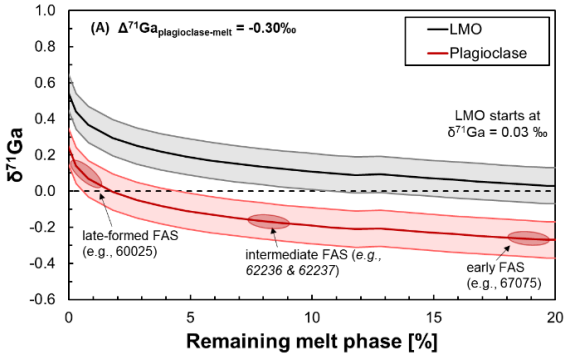

Figure 2: Partitioning of light Ga isotopes into plagioclase with a $\Delta^{71}Ga_{\text{plagioclase-melt}}$ of -0.30‰: These models indicate that the LMO may have started with a Ga isotopic composition that is similar compared to the BSE and indistinguishable from an independent estimate extrapolated from lunar mare basalts [5]. Shaded areas represent 0.10‰ error envelopes on the modeled compositions of the LMO and plagioclase cumulates.

Conclusions: Combining Ga isotope data with trace element abundances reveals that the observed Ga isotope heterogeneities in FAS, MGS, and mare basalt suite rocks were likely controlled by igneous processes during the later solidification stages of the LMO. Using Ga isotope partitioning models, we find that the δ^{71}Ga value of the Moon may be indistinguishable from current estimates for the BSE. Together with constraints from nucleosynthetic and radiogenic isotope systems, these data indicate that the proto-Earth and Theia accreted in similar regions of the volatile-depleted inner Solar System.

References: [1] Canup and Asphaug (2001), *Nature* [2] Day and Moynier (2014) *Philos. Trans. Royal Soc.* [3] Wood et al. (2019) Am. Mineral. [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] McIntosh et al. (2020) *EPSL* [9] Sossi et al. (2018) *PNAS.* [10] Day and Moynier (2014) *Philos. Trans. Royal Soc.* [11] Tartèse et al. (2021) *PNAS.* [12] Kato et al. (2015) *Nature Comms.* [13] Warren (1988) *LPSC XVIII* [14] Sio et al. (2020) *EPSL* [15] Malvin and Drake 1987. [16] Borg et al., (2022) *PNAS* [17] Render and Brennecka (2021) *EPSL* [18] Akram and Schönbächler (2016) *EPSL.*