

LUNAR MAGMA OCEAN CRYSTALLISATION: CONSTRAINTS FROM EXPERIMENTAL AND ANALYTICAL STUDIES. J. F. Snape¹, A. A. Nemchin², M. J. Whitehouse³ and W. van Westrenen¹, ¹Faculty of Science, Vrije Universiteit Amsterdam, the Netherlands (j.f.snape@vu.nl), ²Department of Applied Geology, Curtin University, Perth, Australia, ³Department of Geosciences, Naturhistoriska Riksmuseet, Stockholm, Sweden.

Introduction: Recent studies have provided new insights into the crystallisation ages and initial Pb isotopic compositions of a range of volcanic lunar samples [1-4]. The results of these studies have been used to determine $^{238}\text{U}/^{204}\text{Pb}$ ratios (μ -values) for the magmatic sources from which these rocks were derived, and to place constraints on the starting μ -value of the primordial Moon [1-4]. A range of lunar formation times can be explained by varying the Moon's starting μ -value in order to generate the very radiogenic lunar initial Pb isotope compositions measured in the samples at different rates. For example, if the starting μ -value was very high (>1000), less time is required for the evolution of Pb isotopic compositions, and a young Moon remains a possibility (~4.4 Ga [5-6]). Meanwhile. Meanwhile, if the Moon formed at 4.5 Ga [7-8], less extreme starting μ -values (~450) would be required.

In either case, these estimated μ -values are significantly higher than those typically calculated for terrestrial mantle sources (typically ~8-10 [9-10]). Perhaps more importantly, it is clear that mantle sources with a significant range of μ -values are also required to explain the range of Pb isotopic compositions for different lunar samples. In the case of the mare basalts with the least radiogenic Pb isotopic compositions, source μ -values of a few hundred are predicted [1,3], while KREEP-rich lithologies would require source μ -values of a few thousand [1-2]. Currently, it is not clear at what stage in the early magmatic evolution of the Moon these elevated μ -values were established, or which process(es) could have generated such a wide range of μ -values.

In the past few years, several new experimental studies of lunar magma ocean (LMO) crystallisation have been published [11-13]. These studies aim to replicate the effects of fractional crystallisation in the LMO and provide more detailed information regarding the mineral assemblages and melt compositions formed towards the end of its crystallisation. Here, we combine the results of the Pb isotopic sample analyses and these experimental studies with the current best estimates for the mineral-melt partitioning behaviour of U and Pb [14], to attempt to place new constraints on the initial μ -value of the bulk Moon and identify the stages of LMO crystallisation that could have resulted in the predicted μ -values of the mare basalt and KREEP sources.

Approach: For each stage of LMO crystallisation defined in the experimental studies, the concentration of U and Pb in the residual melt (C_L) is calculated using the Rayleigh fractionation equation:

$$C_L = C_0 \times F^{(D-1)}$$

where C_0 is the concentration of the element in the starting liquid (or the residual liquid of the previous crystallisation stage), F is the melt fraction, and D is the bulk partition coefficient for the minerals generated in that stage. In this instance, the starting μ -value of the LMO has been assumed to be 450. The concentration of U and Pb in the cumulate assemblages (C_S) is calculated as:

$$C_S = C_L \times D$$

One of most significant sources of uncertainty in calculating these values comes from the partitioning behaviour of U and Pb in clinopyroxene (cpx) and, in particular, the uncertainties (stated below as 1σ standard deviations) involved in calculating the cpx partition coefficients for U (D_U) [14]:

$$\ln\left(\frac{D_U}{X_{Ca}^{melt}(X_{Al}^{melt})^2}\right) = -5.64(\pm 1.84) + 6.04(\pm 2.53)(1 - X_{Mg}^{melt})^2 + 5.00(\pm 0.55)(1 - X_{Wo}^{cpx})^2$$

where X indicates the molar fractions of the stated elements in either the residual melt or the cpx (Wo = atomic $\text{Ca}/[\text{Ca}+\text{Mg}+\text{Fe}]$).

Discussion: In all three of the LMO crystallisation studies [11-13], olivine and orthopyroxene are the only phases formed until approximately 70% of the LMO has crystallised. After this, cpx and plagioclase begin to crystallise, resulting in the most significant changes in the μ -values of the residual melt and the mafic cumulates (Fig. 1). Based on the calculations performed here, the current uncertainties in U and Pb partitioning behaviour result in greater variations in these predicted μ -values than any differences between the three previous LMO crystallisation studies. This is most notable with the order of magnitude variations associated with μ -values of the mafic cumulates formed during the final stages of LMO crystallisation (Fig. 1).

μ -value of the residual LMO melt: Despite the potential variation in the partitioning behaviour of U and Pb, and the resulting μ -values of the LMO cumulates, the maximum possible μ -value of the residual liquid in the final stages of LMO crystallisation is less than $10\times$ the starting μ -value. The majority of this increase results from the crystallisation of plagioclase, which acts as a significant Pb sink (Fig. 1). Assuming that the final LMO residual melt composition can be taken as a

proxy for the composition of the “urKREEP” reservoir from which the KREEP-rich basalts obtained their high- μ signatures [1], this provides a rough constraint on the μ -value at the start of LMO crystallisation. As such, if the μ -value of the LMO residual liquid is to reach similar values (~2000-4000) to those determined for the source reservoirs of the KREEP basalts, it is necessary for the LMO to begin crystallising with a μ -value of (at least) several hundred.

μ -values of mafic cumulates: The early mafic cumulates (those comprising olivine and orthopyroxene) have μ -values lower than those of the LMO residual liquid. In general, the crystallisation of cpx, after approximately 70% crystallisation of the LMO, results in an increase of the mafic cumulate μ -values, such that it is possible to generate mafic cumulates with μ -values that are several orders of magnitude larger than those of the LMO residual liquid. As such, a relatively low starting μ -value for the LMO would still be sufficient to produce mafic cumulate μ -values within the range of those previously predicted for the mare basalt sources [1,3] (Fig. 1).

An exception to this is if one set of extreme clinopyroxene D_U values are used, which make it possible to generate mafic cumulates with consistently lower μ -values than the LMO residual liquid. Notably, the potential to generate lower μ -values for the mafic cumulate (relative to the LMO liquid) opens up the possibility of a significantly higher starting μ -value for the LMO and, thus, later formation of the Moon.

Conclusions and future work: The results of these initial calculations demonstrate how sensitive models of lunar magmatic evolution are to uncertainties in partitioning behaviour, and underscore the need for new experimental data to investigate partitioning behaviour of U and Pb in conditions relevant to LMO crystallisation. In spite of these uncertainties, there is still a requirement for the Moon to have acquired a high- μ signature early in its evolution, and prior to the start of LMO crystallisation. However, the current uncertainties preclude placing very precise limits on the starting μ -value, and leave open the possibility for a range of lunar formation times.

A series of new experiments will aim to address these uncertainties by constraining the partitioning behaviour of U and Pb for the same LMO conditions investigated by [11-13]. In addition to this, the partitioning behaviour of other radiogenic parent-daughter pairs will be investigated (including Sm-Nd, Rb-Sr and Lu-Hf), in order to provide better context for interpreting a range of isotopic datasets.

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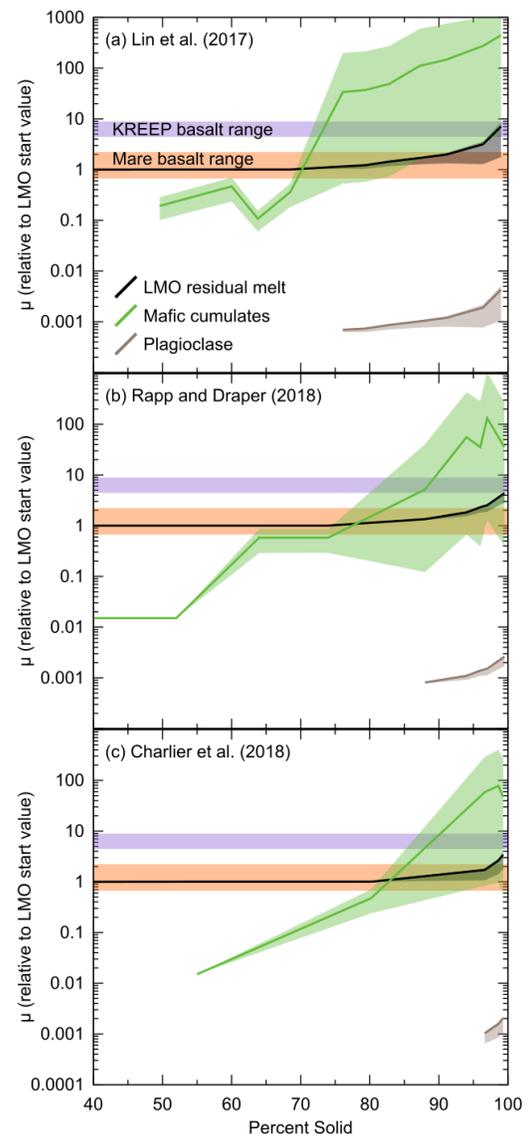


Figure 1: Evolving μ -values the LMO residual liquid, mafic cumulates and plagioclase predicted to form in the experimental studies previously published [11-13]. These are compared with predicted source reservoir μ -values of mare basalts and KREEP basalts [1,3]. The semi-transparent fields surrounding the model μ -values represent the potential range of values resulting from the uncertainties in the partition coefficients of U and Pb.