EUROPIUM MASS BALANCE IN THE LUNAR CRUST (REPEATED);

Randy L. Korotev & Larry A. Haskin, Dept. of Earth & Planetary Sciences and the McDonnell Center for the Space Sciences, Washington Univ., St. Louis, MO, 63130.

Is there an excess of Eu in the polymict samples? Korotev and Haskin [1] argued, on the basis of data for many polymict samples from many missions as well as lunar meteorites that the lunar crust contains an "excess" of Eu (see also [2]). This "excess" is relative to models that derive the Eu of polymict highlands materials from plagioclase with the composition of the plagioclase found in ferroan anorthosites [3]) plus some type of KREEP [4-10]. We showed that polymict samples contain more Eu than allowed by their maximum possible proportions of such plagioclase (based on the Al concentrations of the samples) and KREEP (based on the Sm and Th concentrations). This model assumed that all the non-KREEP plagioclase contains 0.8 ± 0.1 μg/g Eu, typical of ferroan anorthosite from Apollos 15 and Apollo 16. Aware of no selective carrier for Eu other than plagioclase, we argued that the plagioclase in the polymict samples must contain, on average, a Eu concentration of >0.8 μg/g. We noted that plagioclase in magnesian-suite pristine rocks (i.e., Mg-suite norites and troctolites) is richer in Eu than that in most known ferroan-suite pristine rocks (i.e., the ferroan anorthosite suite, FAS [3]), but reaffirmed that the common members of the magnesian suite could not alone provide the mafic components of the polymict samples because their Mg/Fe ratios are too high [1,9,11].

Warren et al. [12] subsequently argued that the Eu "excess" does not exist. Their argument is based on a figure similar to our Fig. 1a, in which only "the most thoroughly polymict" samples, the highlands soils and lunar meteorites, were considered. In Fig. 1, we have added data for the most KREEP-poor soil from Apollo 16 (67511), an Apollo 15 soil (the most aluminous soil, that from the bottom of drive tube 15007), recently discovered lunar meteorite MAC88105, and separate points for meteorites Y82192/3 and Y86032 (which have identical cosmic-ray exposure ages [13], but are nevertheless compositionally distinct) to the set of samples used by Warren et al. [12]. Our original analysis included all of these samples except MAC88105, plus a large number of other polymict samples [1].

Warren et al. [12] argue that the lunar meteorites and the KREEP-poor soils do not have substantially greater concentrations of Eu than found in plagioclase from ferroan anorthosite. This is true, but misses the relevant point. The polymict samples are substantially more mafic (lower Al concentrations) than pure plagioclase. Their comparable Eu concentrations therefore require higher plagioclase Eu concentrations than those of FAS plagioclase, as long as plagioclase is the main carrier of Eu. The differences are shown by the horizontal offset of the polymict sample data from a line connecting typical FAS plagioclase and a hypothetical Eu-free (mafic) diluent [2]. Samples such as those in Fig. 1 containing between 100 and 160 mg/g Al (19 to 30 % Al_2O_3) could have only 0.43 to 0.68 μg/g Eu if all their Eu were in plagioclase with 0.8 μg/g Eu. All the samples in Fig. 1a, including the lunar meteorites, are richer in Eu than that.

Apollo regolith materials contain KREEP, which has both plagioclase and non-plagioclase Eu-bearing components. We also showed that the maximum allowable KREEP (based on Sm and Th concentrations) in the polymict samples is too small to account for the Eu shortfall of a model using plagioclase with 0.8 μg/g Eu as the non-KREEP carrier of Eu, even though KREEP is the principle carrier of Eu in REE-rich polymict samples [1]. We can demonstrate this as follows: The average Sm concentration of the five meteorites plotted in Fig. 1 is 0.9 μg/g. These meteorites are essentially devoid of KREEP [1,14]. Let 0.9 μg/g also represent the concentration of Eu in the non-KREEP portion of Apollo and Luna <1 mm fines; the rest is taken to come from KREEP (48 μg/g Sm [15]), and the proportions of KREEP and non-KREEP can thus be computed. The average Eu concentration of KREEP is 3.3 μg/g [15]. Subtraction of the KREEP Eu from the observed Eu concentration for each polymict sample and normalization to the non-KREEP fraction gives the Eu concentration of that fraction. The resulting "KREEP-corrected" Eu concentrations are plotted in Fig. 1b. This figure shows that even after accounting for KREEP, polymict samples from the lunar highlands contain more Eu than can be explained by plagioclase such as that in Apollo 16 ferroan anorthosite. For the Apollo 14 regolith, much of that excess is probably carried by alkali anorthosites, but that is probably not the case for the other regoliths [1]. Excluding Apollo 14, there is an excess Eu concentration of 48 %, on average, for the regoliths of Fig. 1. Using the Al concentrations of the non-KREEP portions of the samples to estimate their proportions of plagioclase leads to an average concentration of Eu in plagioclase in the non-KREEPy portions of the polymict samples of $0.8 \times 1.48 = 1.18$ μg/g Eu. This is similar to the value previously obtained using a different data set ("1.12-1.16" [1, p. 1805]).

The average plagioclase concentration of ferroan-suite rocks. Warren has long noted that most samples of *pristine* FAS rocks are anorthosites containing $\geq$95 % plagioclase and that many, in fact, contain >98 % [3,16,17]. Although such samples dominate the known FAS, Warren et al. [12] and Warren [3] have criticized our use of compositions representing ferroan anorthosite containing essentially ~100 % plagioclase, such as samples 15415 and 60015, as a component in our model [1]. However, the question our model addressed was the average Eu concentration of lunar crustal plagioclase, and we used monomineralic plagioclase rocks to represent *plagioclase*, not whole rocks.

A principle conclusion of the past decade of our work [1,9,11,18,19] has been exactly the point that Warren has recently recognized, namely, that despite its dominance among samples recognized as "pristine", ferroan anorthosite in its typical pristine form ($\geq$95% plagioclase [9]) cannot be a dominant constituent of the lunar crust; more mafic varieties of FAS rock are required. Korotev and Haskin [1] noted that mass balance constraints for Si, Al, Fe, Mg, Sm, and Eu in the crust could be met with "... a mixture of 51 % ferroan anorthositic norite 67215 [not pristine, but probably a brecciated FAS rock, 20], 21 % ferroan anorthosite 15415, 23 % magnesian norite 78235, 1 % magnesian troctolite 76535, and 3.8 %

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KREEP..." This is simply one of a number of possible solutions that depend on the compositions of the assumed components. We were careful not to imply that literally two ferroan lithologies were required, one containing ~100% plagioclase (15415 type) and another containing 68% plagioclase (67215 type), but that FAS components overall accounted for ~72 wt% of the crust and that the average plagioclase concentration of those FAS components was ~78 wt% (~81 vol%), i.e., that of anorthositic norite, not of anorthosite. Here we use "FAS" to represent a probable variety of primitive rocks with relatively low Mg# (molar Mg/[Mg+Fe] ratio), high An, and low Eu concentrations, not necessarily limited to the pristine ferroan-suite rocks reported in the literature so far. When we stated that "...probably less than half [of the upper crust] is *ferroan anorthosite* or brecciated derivatives" [1], we meant *ferroan anorthosite* of the Apollos 15 and 16 type containing >90 vol% plagioclase [21]. As in the mass-balance example above, FAS rocks are an important constituent of the crust, but more mafic varieties are more important than rocks like the Apollo 16 ferroan anorthosites. We have repeatedly shown that noritic anorthosite or anorthositic norite lithologies must have been major carriers of mafic minerals of the early lunar crust, their rarity as pristine rocks notwithstanding, and that the mafic components must be a balance between primitive magnesian and ferroan materials such that the resultant Mg# is ~0.7, similar to highlands soils [1,9,11]. The meteorites from the lunar highlands are regolith breccias that are typical of the highland surface crust [18,22]. These meteorites contain primitive clasts that span a wide range of Mg# and that are generally more mafic than typical pristine FAS rocks from Apollo 16 [18,22]. This supports the conclusions of our modeling [1] that more mafic FAS rocks are a major, but cryptic, component of the Apollo polymict samples and that the pristine FAS rocks at Apollo 16 are atypically rich in plagioclase [23].

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Fig. 1. Average aluminum and europium concentrations in highlands regoliths, after [12] and [2]. The triangle represents the concentrations (35.5 % Al_2O_3, 0.8 ± 0.1 μg/g Eu) in plagioclase from typical ferroan anorthosite from Apollo 16 and Apollo 15 (15415) [1]. The dashed line represents the effect of "dilution" with the other principle mineral constituents (pyroxenes, olivines, opaques). Raw data are plotted in (a). In (b), the concentrations of both elements have been corrected for KREEP based on the Sm concentrations of the regoliths and the concentrations of Al, Eu, and Sm in KREEP [15] (see text). Fig. 1b estimates the concentrations in the nonKREEP portion of these polymict samples. The figure shows that the plagioclase of the lunar surface crust has an average Eu concentration about 50 % greater than the concentration in plagioclase of typical ferroan anorthosite. This "excess" is most likely contained in magnesian-suite (Mg-suite) norites, troctolites, gabbros and their more anorthositic variants; for Apollo 14 soil, some is probably contained in alkali anorthosites, hence the large excess [1]. Regolith data are from the following sources: Apollo 14 [9], Apollo 15 (least contaminated with mare basalt) [24], Apollo 16 (typical) [9], 67511 (Apollo 16, lowest REE concentrations) [25], Apollo 17 (least contaminated with mare basalt) [9], Luna 20 [9]; lunar meteorites (means): ALHA81005 [26], Y-791197 [27], Y-82192/3 [28], Y-86032 [24], MAC88105 [20].