INTERNAL LU-HF ISOTOPE SYSTEMATICS OF THE EUCRITES MILLBILLILLIE AND PIPLIA KALAN. R. Bast[1], E. E. Scherer[1], S. Taetz[1], P. Sprung[2], K. Mezger[3] and G. Srinivasan[4], [1]Institut für Mineralogie, Westfälische Wilhelms-Universität Münster, Corrensstr. 24, D-48149 Münster, Germany, Rebecca.Bast@uni-muenster.de, [2]Institut für Geochemie und Petrologie, Eidgenössische Technische Hochschule Zürich, Clausiusstr. 25, CH-8092 Zürich, [3]Institut für Geologie, Universität Bern, Baltzerstr. 1+3, CH-3012 Bern, Switzerland, [4]Center for Earth Sciences, Indian Institute of Sciences, Bangalore.

Introduction: The high-precision absolute dating of early solar system materials and events currently relies on the U-Pb system. Another potentially precise chronometer for such applications is the Lu-Hf system, whose high closure temperature in silicates [1] and phosphates [2] should make it relatively robust against later disturbances such as thermal metamorphism and impacts. However, the Lu-Hf ages of meteorites are often > 100 Myr older than their U-Pb (Pb-Pb) counterparts, and thus much older than the Solar System age defined by CAI (4568.2 Ma [3]). For example, the trend defined by eucrite whole rocks implies an age of ca. 4.71 Ga (data of [4, 5, 6]), or 3% too high. Here, we investigate the possible causes of the 2-5% age discordance and present new Lu-Hf internal isochron data that illuminate complexities within eucrite samples.

Possible causes of the age discrepancy: These include: sample complexities (i.e., isochron assumptions not met) and analytical artifacts. We do not consider the accuracy of the ^{176}Lu decay constant (λ^{176}Lu) to be a contributing factor because its determination by age comparison against the U-Pb system yielded a precise value of $[1.867 \pm 0.011] \times 10^{-11}$yr^{-1} (2 s.d.) for 9 different terrestrial samples ranging in age from 0.9 to 2.8 Ga [7, 8, 9]. These samples were selected specifically for the purpose of determining λ^{176}Lu, i.e., they all stemmed from intrusive rocks that 1) were precicely datable by both U-Pb and Lu-Hf, 2) cooled rapidly enough to minimize the effects of closure temperature differences among minerals, and 3) have not been significantly disturbed since crystallization. In contrast, many meteorites are characterized by complexities such as brecciation, shock metamorphism, and cosmic ray exposure, that preclude their use in decay constant calibrations. Nevertheless, some recently measured internal Lu-Hf isochrons of meteorites or their components yielded reasonable ages, [e.g., 10, 11, 12, 13] and this has been taken as evidence that λ^{176}Lu has been indeed constant since the start of the solar system [10].

Significant analytical artifacts can be ruled out as the cause of the spurious Lu-Hf ages because laboratories using different chemical separation methods, independently calibrated spikes, and different mass spectrometers report similar slopes and intercepts for Lu-Hf isochrons of eucrites and chondrites [e.g., 4, 5, 6].

Because the decay constant and analytical methods cannot explain the high Lu-Hf ages, sample complexities, including the ones previously mentioned, should be explored. It has been proposed that irradiation of early solar system materials by either gamma rays [14] or cosmic rays [15] could have produced the short-lived isomer ^{176m}Lu ($t_{1/2}$ = 3.7 hrs), accelerating the ^{176}Lu-decay, and leading to an excess of ^{176}Hf proportional to a sample's Lu/Hf. Accordingly, internal isochrons for differentiated meteorites would rotate, increasing the apparent age without producing scatter or affecting the initial ^{176}Hf/^{177}Hf.

Samples: To test whether eucrites meet isochron assumptions and to further test for irradiation-induced excess ^{176}Hf, we investigated the monomict basaltic main-group eucrites Millbillillie (MB) and Piplia Kalan (PK). Both are witnessed falls and thus not expected to have been influenced by terrestrial weathering. Further, they both show evidence of live ^{26}Al, indicating that they crystallized within 5 Myr of CAI formation [16, 17]. An absolute age of 4564 Ma has been calculated for MB from its excess ^{26}Mg [17], correlating well with the reported Lu-Hf age of 4566 ± 93 Ma [11]. For PK, a Sm-Nd age of 4570 ± 23 Ma has been reported [18, 19].

Methods: An 8 g piece of MB and about 2 g of PK were crushed and separated into different density and magnetic fractions and then handpicked. Dry aliquots of mineral fractions were reserved for later Pb-Pb dating, with the remainders being spiked for Lu-Hf and Sm-Nd, then digested in HF-HNO_3-$HClO_4$. Pure px and plag digestions were carried out on a hotplate (to avoid digestion of any zircon inclusions) and in high pressure autoclaves for wr and ilmenite (to ensure complete sample digestion including any zircon). Lutetium and Hf were separated by ion exchange chemistry as described in [20] and measured by MC-ICP-MS at the Institut für Mineralogie in Münster. The major- and trace-element-bearing matrix was kept for additional Sm-Nd isotope analyses.

Results: Internal "errorchrons" for MB and PK reveal a large degree of scatter, i.e., neither meteorite yielded a single, isochronous trend among all its mineral- and bulk fractions. Individual data points typically plot above the eucrite whole-rock (wr) trend defined by data from [4, 5, 6] (Fig. 1), with plagioclase (plag) being the most apparently disturbed fraction. Similar features have been observed in the eucrite NWA 5073 [21]. The pure pyroxene (px)-wr of both MB and PK trends yield apparent ages of ca. 4.9 Ga (using λ^{176}Lu $= 1.867 \times 10^{-11}$ yr^{-1}) with an apparent initial $^{176}Hf/^{177}Hf$ of 0.27959(5) for MB and 0.27957(6) for PK. That the ages and $^{176}Hf/^{177}Hf_i$ are higher and lower, respectively, than for the eucrite whole rock trend (e.g., 4.71 Ga, 0.27970(3); data of 4, 5, 6]) suggests that px fractions have also been disturbed. A second trend represented by the wr, composite, and plagioclase fractions of MB, yields an apparent age of 3.70(6) Ga (Fig. 2).

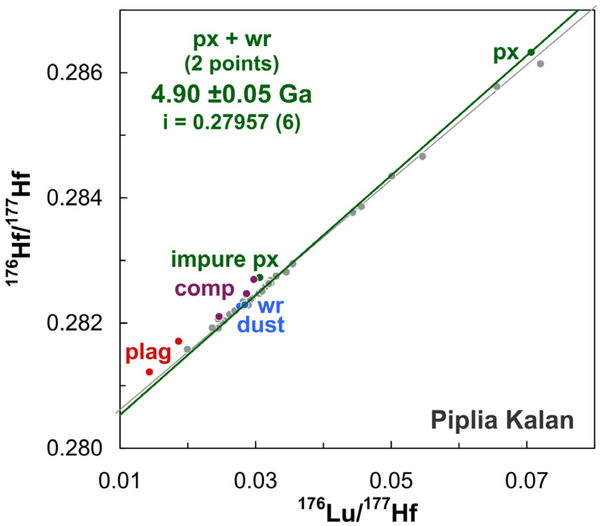

Fig. 1: Lu-Hf isotope data for different fractions of Piplia Kalan, plotted on the eucrite whole-rock (wr) trend (grey) defined by the data of [4], [5], and [6]. Note that most mineral fractions plot above the eucrite wr isochron, whereas PK's wr and dust fractions plot on this trend.

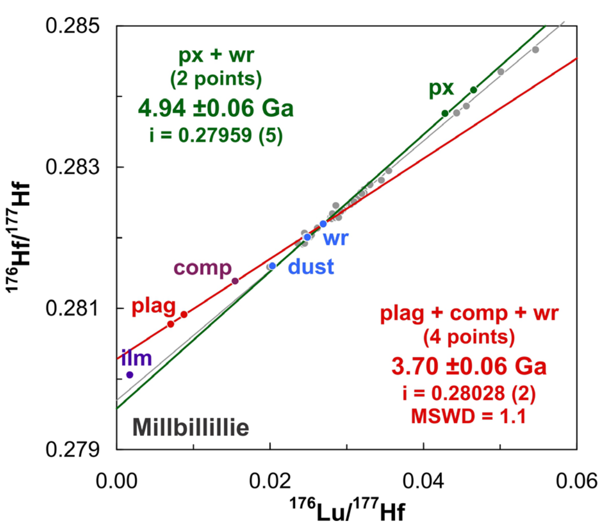

Fig. 2: The internal Lu-Hf isotope data for Millbillillie appear to define two different trends rather than a single isochron.

Interpretation: The observed internal complexities indicate a partial redistribution of Lu and/or Hf among the different constituent minerals. Nevertheless, both eucrite whole rocks appear to have remained essentially closed systems with respect to Lu and Hf because they both lie on the eucrite whole rock trend. Although low-Hf phases such as plag should be more susceptible to the influence of Hf released from other minerals during disturbance, even the purest px fractions also seem to have been affected. The relationship of the wr samples to hot-plate digested samples could indicate that an ancient resetting event occurred in which one or more minerals (e.g., zircon) did not participate fully in the isotopic equilibration. Such "inherited" zircon would pull the wr below the trend defined by the zircon-free fractions [1]. However, such zircon would have to have a lower $^{176}Hf/^{177}Hf_i$ than implied by the eucrite wr trend, which seems unlikely.

We conclude that with the current data set, no meaningful isochron slopes can be determined and we cannot yet exclude the possibility of accelerated ^{176}Lu decay via irradiation in these samples.

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