Hf-W ISOCHRON FOR BULK CAI: EVIDENCE FOR HOMOGENEITY OF ^{26}Al AND ^{182}Hf

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Introduction: The timespan between condensation of the first solids and melting and differentiation of the first planetary bodies probably was only a few Myr. Establishing a fine-scale chronology of early solar system evolution, therefore, requires the determination of highly precise ages for meteorites and their components. The necessary time resolution is in principle provided by Pb-Pb chronometry and by short-lived chronometers such as the ^{26}Al-^{26}Mg and ^{182}Hf-^{182}W systems. However, the precise chronological interpretation of the isotopic data is not always straightforward, and can be hampered by (*i*) mass-independent isotope variations in the elements of interest [*e.g.,* 1-3] and (*ii*) a heterogeneous distribution of some short-lived radionuclides (especially ^{26}Al) [*e.g.*, 4].

In a previous study we have shown that the interpretation of Hf-W data for CAI is complicated by the presence of nucleosynthetic W isotope anomalies [1]. To explore the extent and nature of nucleosynthetic W isotope variations in CAI, and to better quantify the initial Hf and W isotope compositions of the solar system, we obtained Hf-W data for several fine- and coarse-grained CAI from different CV3 chondrites. The improved initial ^{182}Hf/^{180}Hf is then used to compare Hf-W and Al-Mg ages between angrites and CAI, with the ultimate goal to assess the level of ^{26}Al heterogeneity in the early solar system.

Methods: After digesting the CAI samples in HF-HNO_3-$HClO_4$, W was separated using two anion exchange chromatography steps modified after [5]. Tungsten isotope compositions were measured on a ThermoScientific® Neptune *Plus* MC-ICPMS at the University of Münster. The data are reported as ε^{i}W (*i.e.*, 0.01 % deviations from terrestrial values) and for two normalizations, denoted '6/3' for normalization to ^{186}W/^{183}W and '6/4' for normalization to ^{186}W/^{184}W.

Results: The *fine-grained* CAI exhibit variable ε^{183}W (6/4) with excesses of up to +5.3, indicating a deficit in *s*-process (or an excess in *r*-process) W nuclides in some of the samples. In contrast, all of the *coarse-grained* (mostly type B) inclusions show near-terrestrial ε^{183}W (6/4), indicating that nucleosynthetic W isotope heterogeneity is largely absent in these samples. Moreover, the investigated CAI exhibit variable ε^{182}W (6/4), while the ε^{182}W (6/3) values of the same samples show a smaller range. This difference results from the presence of nucleosynthetic W isotope anomalies in some of the CAI, which have a much larger effect on ^{186}W/^{184}W-normalized than on ^{186}W/^{183}W-normalized ratios (Fig. 1).

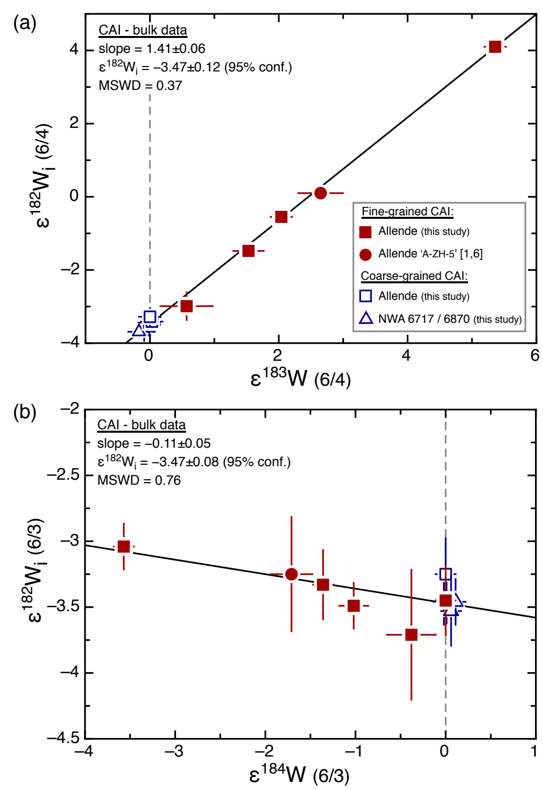

Fig. 1: Tungsten isotope systematics of bulk CAI normalized to (a) ^{186}W/^{184}W, and (b) to ^{186}W/^{183}W after iteratively correcting the measured ε^{182}W of each CAI for ^{182}Hf decay using their measured ^{180}Hf/^{184}W.

Effect of nucleosynthetic W isotope anomalies on Hf-W systematics: The fine-grained CAI investigated here display a sufficiently large range in Hf/W to permit determining a precise bulk CAI isochron. However, the ε^{182}W values of the CAI first must be corrected for nucleosynthetic anomalies, which requires knowledge of the relative effects of such anomalies on ε^{182}W and ε^{183}W (or ε^{184}W). This information is provided by the slope of ε^{182}W *vs.* ε^{183}W (or ε^{184}W) correlation lines. After an iteration to correct the measured ε^{182}W of each individual CAI for radiogenic contributions from ^{182}Hf decay, the studied fine-grained CAI define a precise correlation between initial ε^{182}W and ε^{183}W (Fig. 1), providing a direct empirical means to correct the ε^{182}W of any CAI for nucleosynthetic isotope anomalies using their measured ε^{183}W.

Hf-W isochron for bulk CAI: After the iterative correction for nucleosynthetic W isotope heterogeneity, a well-defined Hf-W isochron for bulk CAI is obtained. Linear regression of the ^{186}W/^{183}W-normalised data provide an initial $^{182}Hf/^{180}Hf$ of $(1.02\pm0.05)\times10^{-4}$ and $\varepsilon^{182}W_i$ (6/3) = −3.48±0.11 (Fig. 2). Both values are in good agreement with a previous determination based on internal Hf-W isochrons for type B CAI [1,6], but the nominal initial $^{182}Hf/^{180}Hf$ obtained for CAI here is slightly higher than that obtained previously. Based on Al-Mg chronometry it was argued that re-melted, coarse-grained CAI may have formed up to ~0.2 Ma later than primitive, unmelted CAI [7]. Although such a brief time interval cannot be resolved using Hf-W chronometry, it would be consistent with the slight (but not resolved) difference between the initial $^{182}Hf/^{180}Hf$ of the bulk CAI isochron and that of an internal isochron for type B CAI [1,6]. The initial $^{182}Hf/^{180}Hf$ of the solar system, therefore, is best determined by the bulk CAI isochron obtained in the present study.

Further evidence that the bulk CAI isochron provides the initial $^{182}Hf/^{180}Hf$ at the time of CAI formation comes from the Hf-W isochron itself, which yields an absolute Hf-W age for CAI of 4567.9±0.8 Ma when anchored to the angrite D'Orbigny [2,5]. This absolute Hf-W age is in good agreement with Pb-Pb ages of CAI [8,9], indicating that the bulk CAI isochron reflects the time of Hf/W fractionation during CAI formation.

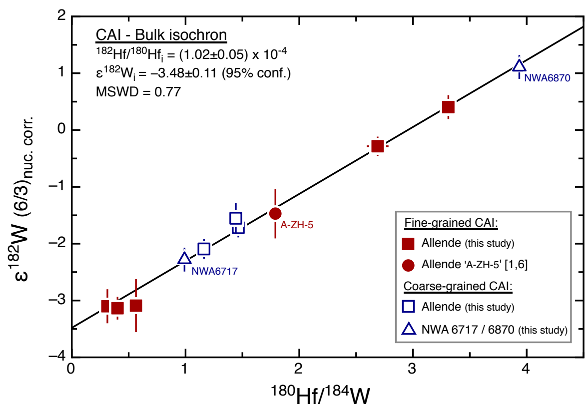

Fig. 2: Hf-W isochron for bulk CAI after an iterative correction of $\varepsilon^{182}W$ for nucleosynthetic W isotope variability.

Evidence for ^{26}Al homogeneity: The use of extinct short-lived chronometers like ^{26}Al-^{26}Mg and ^{182}Hf-^{182}W relies on the assumption that the parent nuclides were homogeneously distributed in the early solar system. However, it has been suggested that variable $^{26}Al/^{27}Al$ in meteorites and meteorite components reflect a heterogeneous distribution of ^{26}Al and not different formation times. For instance, [4] proposed that the angrite parent body derived from a reservoir with initial $^{26}Al/^{27}Al$ of $(1.61\pm0.32)\times10^{-5}$ at the time of CAI formation. Relative to this value the Al-Mg age for the angrites D'Orbigny and Sahara 99555 would be 3.5±0.2 Ma. This is inconsistent with the Hf-W age of these two angrites [5] of 4.8±0.6 Ma relative to the initial $^{182}Hf/^{180}Hf$ of CAI inferred in the present study (Fig. 3). Thus, our new Hf-W data do not support the heterogeneous distribution of ^{26}Al inferred by [4]. Instead, the Hf-W age of the angrites is in excellent agreement with their Al-Mg ages of 4.7±0.2 Ma [10] or 5.0±0.1 Ma [11] relative to the canonical initial $^{26}Al/^{27}Al$ of CAI [12] (Fig. 3). This provides powerful evidence that ^{26}Al (and ^{182}Hf) were homogeneously distributed in the inner solar system, at least in the region were CAI and angrites formed.

Fig. 3 also shows Pb-Pb formation intervals for angrites, calculated relative to two different Pb-Pb ages of CAI, both of which have been calculated using measured U isotope compositions [8,9]. These two Pb-Pb ages for CAI differ by ~0.6 Myr, somewhat hindering the precise determination of Pb-Pb formations intervals of angrites relative to CAI. This makes it difficult to use Pb-Pb ages for assessing the level of ^{26}Al heterogeneity by age comparison. Fig. 3 shows that the Hf-W data seem more consistent with the older Pb-Pb age of CAI, but the Hf-W ages do not have the required precision to unambiguously distinguish between the two different Pb-Pb ages for CAI.

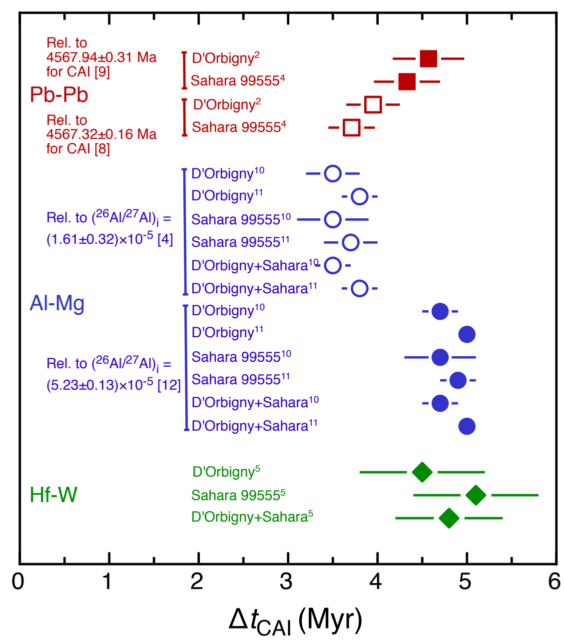

Fig. 3: Hf-W, Al-Mg, and (U isotope variability-corrected) Pb-Pb ages of angrites relative to CAI formation.

References: [1] Burkhardt C. et al. (2012) *APJL* 753, L6. [2] Brennecka G.A. & Wadwha, M. (2012) *PNAS* 109, 9299-9303. [3] Wasserburg G.J. et al. (2012) *MAPS* 47, 1980-1997. [4] Larsen K.K. et al. (2011) *APJL* 735, L37. [5] Kleine T. et al. (2012) *GCA*. 84, 186-203. [6] Burkhardt C. et al. (2008) *GCA* 72, 6177-6197. [7] MacPherson G.J. et al. (2012) *EPSL* 331, 43-54. [8] Connelly J.N. et al. (2012) *Science* 338, 651-655. [9] Bouvier A. et al. (2011) Workshop on Formation of the First Solids in the Solar System, #9054. [10] Spivak-Birndorf L. et al. (2009) *GCA*. 73, 5202-5211. [11] Schiller M. et al. (2010) *GCA*. 74, 4844-4864. [12] Jacobsen B. et al. (2008) EPSL 272, 353-364.