TUNGSTEN NUCLEAR ANOMALIES IN IRON METEORITES AND IMPLICATIONS FOR Hf-W CHRONOLOGY.

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Introduction: The extinct ^{182}Hf-^{182}W chronometer ($t_{1/2}$=8.9 My) is useful to date processes of planetary differentiation, and in particular metal-silicate segregation in the early solar system [1-6]. However, use of this chronometer rests on the assumption that the solar system was an originally homogeneous reservoir in terms of its W isotopic composition. Planetary-scale isotopic variations have been documented for noble gases, O, Cr, Mo, Ru, Ba, Sm and Nd [e.g., 7-12]. For heavy elements, these variations may reflect incomplete mixing of products of stellar nucleosynthesis in the solar nebula. Similar anomalies, albeit of larger magnitude, have been found in presolar grains.

Tungsten has 5 isotopes of which only 4 (182, 183, 184 and 186) can be measured with high precision (180 has very low abundance). The ^{186}W/^{183}W pair is used for internal normalization. Thus only one isotope, ^{184}W, is left beyond ^{182}W to quantify the degree of mixing of nucleosynthetic products in the solar nebula.

Methods and Materials: We measured the W isotopic compositions of iron meteorites from groups IA-IIICD, IIE, IC, IIAB, IID, IIIAB, IIIE, IIIF, IVA, IVB and one ungrouped iron, Deep Springs, using a GV Instruments Micromass Isoprobe multicollector inductively coupled plasma mass spectrometer (MC-ICPMS) after chemical separation by ion chromatography, at the Field Museum (see ref. [13] for details). The same batches of solutions of Cedartown (IIAB), Tlacotepec (IVB) and Deep Springs used for Isoprobe analyses were also analyzed by Neptune MC-ICPMS at the Thermo Factory in Bremen. The same samples were also processed with a different chemical protocol and analyzed by Nu-plasma MC-ICPMS at ETH (see [6] for details of the method).

Results: The results are reported in ε units (i.e., the deviation of internally-normalized W isotopic ratios relative to a terrestrial standard × 10^4). The ε^{182}W (ε^{182}W/^{183}W) results agree with previous work [6] and will not be discussed hereafter. Most iron meteorites show ε^{184}W(ε^{184}W /^{183}W) values identical to the terrestrial standard within 95% confidence intervals. Within each group, no variation was observed for ε^{184}W outside the analytical error. However all the meteorites from group IVB show systematic deficits in ε^{184}W with a group mean of -0.08±0.01. A larger deficit in ε^{184}W was observed for Deep Springs with a mean of -0.15±0.02 for 5 analyses. The measured ε^{184}W values for IIAB, IVB and the Deep Springs iron meteorites are shown in Fig. 1.

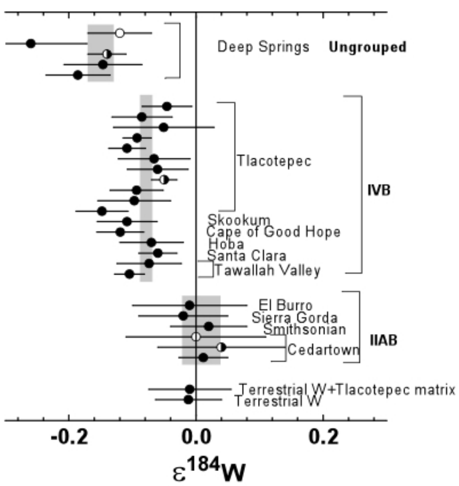

Fig. 1. ε^{184}W for IIAB, IVB, and the Deep Springs iron meteorites and terrestrial standards. Solid symbols, half-open and open symbols correspond to measurements obtained by Isoprobe, Neptune and Nu-plasma MC-ICPMS, respectively. The error bars are 95 % confidence intervals. The gray areas show the weighted averages and corresponding error bars for each group.

Discussions: *Accuracy of the method.* Because the deficits we observed for IVBs and Deep Springs are at the 0.1 ε level, care must be taken to ensure that these are not caused by analytical artifacts, such as (i) effects due to matrix elements and interfering isobars in the analyzed solutions, (ii) inappropriate correction of mass fractionation, (iii) mismatch in W concentration between the sample and bracketing standard. We tested all these factors and concluded that they cannot be the cause for the measured deficiencies in ε^{184}W. More importantly, there is excellent inter-laboratory agreement between Chicago, Bremen, and Zurich for the three iron meteorites that have been studied in more detail (Fig. 2).

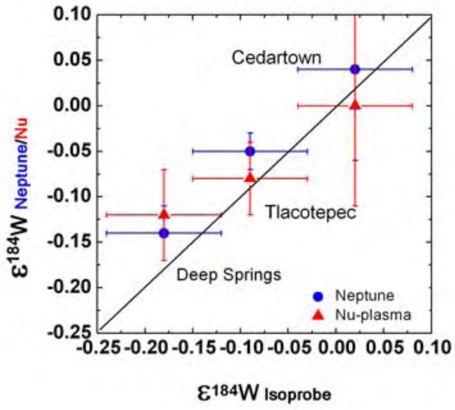

Fig. 2. ε^{184}W values obtained for Cedartown, Deep Springs and Tlacotepec using three different types of MC-ICPMS (Isoprobe, Neptune and Nu plasma). All data are from this study, except Tlacotepec ε^{184}W by Nu-plasma from [6].

Cosmogenic effects? Negative anomalies in ε^{182}W lower than the inferred solar system initial value [1, 14] have been documented for some iron meteorites including Tlacotepec, and was explained by exposure to galactic cosmic rays in space [15-17]. However GCR irradiation cannot account for the deficiencies in ε^{184}W of IVBs. Indeed, if this was the case, a correlation would be expected between ε^{184}W and ε^{182}W for IVBs. In addition, the correlation line should pass through ε^{184}W=0, when ε^{182}W=-3.5 (assuming that the original ε^{182}W value of IVBs before GCR exposure is close to the initial value inferred for the solar system [1, 14], as documented for other iron meteorite groups [6]). Our data show no correlation between ε^{184}W and ε^{182}W and ε^{184}W~-0.1 when ε^{182}W=-3.5.

Nucleosynthetic effects. The most likely explanation for the deficiencies in ε^{184}W measured in IVBs is that they reflect heterogeneous distribution of the products of stellar nucleosynthesis in the solar nebula. All major W isotopes are synthesized by *s*- and *r*-processes. Using an updated nuclear reaction network [18], we computed the *s*-process yields of W isotopes in an AGB stellar model. A deficiency in ε^{184}W can be explained by a slight deficiency in *s*- or enrichment in *r*-process component. *s*-component deficiencies in iron meteorites have also been documented for Mo [9] and Ru [10]. The W isotopic anomalies recorded in the IVB irons must reflect heterogenous distribution of *s*- and *r*-process nuclides at the scale of the parent-body of these meteorites (~2-4 km in radius [19]), since the carrier phases of nucleosynthetic anomalies must have been destroyed during melting and segregation of metallic cores.

Mixing of *s*-process W with terrestrial composition predicts that there should be a correlation between non-radiogenic, non-cosmogenic ε^{182}W and ε^{184}W (both normalized to ^{186}W/^{183}W) with a slope of 0.036. Thus, a deficit of ~0.1ε in ε^{184}W translates into a minor correction of 0.0036 ε in ε^{182}W, which is negligible compared to the analytical error and no correction needs to be made to the model age (metal/silicate differentiation age). However, the correction slope is uncertain due to the large uncertainties of some key nuclear properties used in the AGB stellar model, namely, the cross sections and beta-decay rates of branching points ^{182}Ta, ^{181}Hf [20] and ^{185}W. We investigated the effect of changing these parameters within their uncertainty ranges and found that it leads to an uncertainty of ~± 0.2 in the correction slope. The maximum correction that needs to be made to model ages remains low (< ± 0.2 My). However a recent study [20] found that the *r*-residue of ^{182}W (by subtracting *s* abundance from solar abundance) shows up as a positive anomaly above the smooth curve of *r*-process abundances, implying that the *s*-process yield of this nuclide may have been overestimated. Changing the cross sections of some branching points does not help in this respect. The problem can be easily solved by lowering the cross section of ^{182}W by 20%. In that case, the calculated slope between non-radiogenic, non-cosmogenic ε^{182}W and ε^{184}W is 0.46; For a deficit of -0.08 ε in ε^{184}W, a correction of ~0.4 My would need to be made to the model age of IVBs. As suggested elsewhere [20], all the cross sections of W isotopes need to be remeasured to better constrain *s*-process abundances and the value of the correction that must be applied to the measured ε^{182}W for model age calculations.

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