TUNGSTEN-180 ANOMALIES IN IRON METEORITES REFLECT ALPHA DECAY OF OSMIUM-184.

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Introduction: Tungsten-180 is among the rarest stable nuclides in the solar system and is mainly produced by the p-process during nucleosynthesis [1]. Precise measurements of the isotopic abundance of ^{180}W in natural materials have long been hampered by the low relative abundance (~0.12%), but recent advances in ICP mass spectrometry have permitted measurements at precisions better than one part in 10,000, as was demonstrated by [2]. In the latter work, positive isotope anomalies of up to 600 ppm were resolved for magmatic iron meteorites relative to the terrestrial composition. Amongst other models, these anomalies were initially interpreted to possibly reflect nucleosynthetic heterogeneity of ^{180}W, i.e., incomplete mixing of a p-process component in the solar system at the time at which the iron meteorite parent bodies formed. This interpretation has however been challenged by the following observations: First, a variability of ^{180}W has recently been reported between individual IVB group iron meteorites [3]. Secondly, the ^{180}W values reported for the Cape York meteorite between different studies [2,4] vary outside the reported analytical uncertainties. Secondary neutron capture on ^{180}W due to cosmic ray exposure may explain some of the observed variability. For Cape York, however, these effects are known to be minimal for ^{182}W (< 10 ppm [5]). Consequently, neutron capture effects on ^{180}W, which has a similar neutron capture cross section [6] should be negligible. It is therefore possible that the ^{180}W heterogeneities in iron meteorites reflect a process different from nucleosynthetic heterogeneity and secondary neutron capture.

Here, we propose that ^{180}W heterogeneities in magmatic and non-magmatic irons can be explained by in situ radiogenic production due to alpha decay of ^{184}Os. Osmium-184 is the rarest nuclide in the solar system in ground state, and is a p-process only isotope [1]. From its nuclear structure ^{184}Os is expected to be unstable, but alpha-particle counting experiments have not confirmed radioactive decay [7]. Therefore, ^{184}Os is commonly regarded as an observationally stable nuclide with a minimum half life of more than 5.6×10^{13} years. Based on this minimum half life it was consequently inferred by [2] that the radiogenic contribution of ^{180}W from ^{184}Os-decay would be negligible for most iron meteorites. However, theoretical estimates of the half life of ^{184}Os range as low as 5×10^{9} years (e.g., [7]), at which the production of ^{180}W would indeed be significant. We therefore performed combined ^{180}W and Os-W concentration measurements to further evaluate the putative alpha decay of ^{184}Os.

Analytical strategy: Eleven iron meteorite specimens from six different classes (IAB, IC, IIAB, IIIAB, IVA, IVB) were selected to cover virtually the whole range of Os/W ratios known from iron meteorites. The cosmic ray exposure ages reported in literature are ≤ 500 Myrs for nine and 700-1000 Myrs for two of these meteorites (refs in [2]). Importantly, the iron meteorite parent bodies are well known to have differentiated within a few Myrs from one another (e.g., [5,8]), a time interval that is negligible compared to the expected half life of ^{184}Os. An isotopic tracer enriched in ^{190}Os was added to the samples prior to digestion in inverse aqua regia using an Anton Paar high-pressure asher. Osmium was subsequently separated by solvent extraction and further purified by microdistillation, after which the Os fractions were measured on Faraday cups by N-TIMS at Berlin. A tracer enriched in ^{183}W was added to a small aliquot of the remaining solution from which W was separated by anion exchange chromatography. The isotope composition of the spiked W fraction was measured by multicollector (MC)-ICPMS. Unspiked W was separated from the main mass of the remaining solution and was used for isotope abundance measurements using the Neptune MC-ICPMS at Cologne-Bonn identical to the procedure described in [2]. The external reproducibility (2σ) for ^{180}W/^{184}W of a 50 ppb terrestrial W-standard solution (AMES metal) was typically better than 65 ppm.

Results: Our samples exhibit variable anomalies of ^{180}W that correlate significantly with Os/W ratios (MSWD = 1.4) (Fig.1). Our data indicate that samples from the same meteorite class (i.e. the IIAB and IVB groups) may have different ^{180}W anomalies, even at low exposure ages where neutron capture is expected to be negligible. Furthermore, we obtain different isotope abundances of ^{180}W for three out of nine meteorites (Cape York, Weaver Mountains, Hollands Store) that were also analyzed by [2], confirming possible variability at the meteorite specimen scale. No inverse correlation is observed between ε^{180}W and W concentration.

Discussion: We propose that the regression shown in Fig. 1 represents an isochron for the iron meteorite parent bodies. This isochron is the first empirical evidence for alpha decay of ^{184}Os. The slope m = 0.000298(61) corresponds to λ^{184}Os(α) = 6.53 ± 1.34 × 10^{-14} a^{-1}, assuming an average age of the iron meteorite parent bodies of 4.565 Ga. The calculated half life of 1.11 ± 0.23 × 10^{13} year is only slightly less than the lower limit that was estimated by alpha-particle counting [7]. Notably, the absence of an inverse correlation between ε^{180}W and W concentration demonstrates that the correlation between ε^{180}W and Os/W ratios cannot be explained by mixing of two compositionally distinct reservoirs with respect to W.

Because all samples plot within error of the fit, the decay by ^{184}Os is evidently the major process explaining the observed ^{180}W anomalies. Therefore, nucleosynthetic heterogeneities of ^{180}W in the samples need to be at the 1ε-unit scale or lower. This is also the case for the two IVA meteorites that appear to have elevated ^{180}W, but overlap the isochron within error. Likewise, nuclear burnout of ^{180}W due to cosmic ray exposure has to be within the limits of analytical precision for the analyzed meteorites. Importantly, also the Bendego IC iron plots within error of the regression, although this meteorite is known to have the longest cosmic ray exposure age in our sample suite, i.e. 940

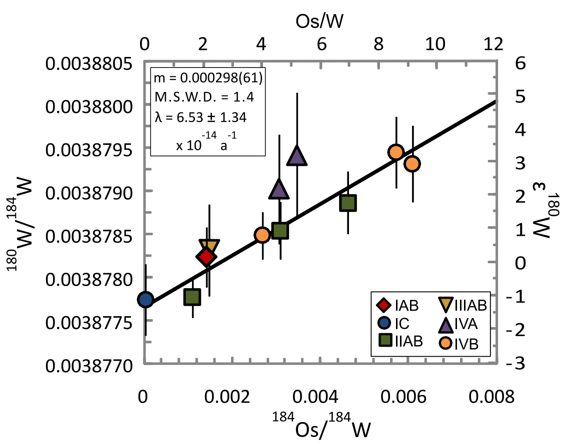

Fig. 1. Measured isotope abundances of ^{180}W plotted against Os/W ratios for 11 iron meteorites covering 6 different classes. Epsilon ^{180}W values are expressed as the deviation from the AMES terrestrial W standard, i.e., $\varepsilon^{180}W = \{[(^{180}W/^{184}W)_{sample}/(^{180}W/^{184}W)_{AMES}]-1\}\times10^4$. A terrestrial ^{180}W/^{184}W composition of 0.0038780 ± 0.0000002 (2σ) used here is the average value obtained throughout the different measuring sessions, normalized to a ^{186}W/^{184}W of 0.92767 using the exponential law. ^{184}Os/^{184}W ratios were calculated from Os-W concentrations, using the atomic weights and isotopic abundances recommended by IUPAC. Errors on ε^{180}W correspond to 2σ r.s.d.. Propagated errors on the ^{184}Os/^{184}W ratios are smaller than symbol sizes.

±70 Myrs [9]. Thus, in contrast to strong neutron capture effects on ^{182}W for this meteorite [2], the effects on ^{180}W appear negligible within analytical uncertainty. Nucleosynthetic and cosmogenic effects on ^{184}Os would have to be in the order of one percent for samples with the largest ^{180}W anomalies in order to affect the isochron significantly. Such strong ^{184}Os effects have been reported for leachates of the Murchison carbonaceous chondrite [10] but are rather unlikely at bulk rock scale. Our data therefore also suggest a homogeneous ^{184}Os distribution at the percent level among different iron meteorite parent bodies, in agreement with [11].

In the context of the proposed decay system, the variability of ^{180}W within iron meteorite groups is explained by variations of Os/W ratios. The Os/W variations are most likely controlled by Os contents that are known to be variable due to solid metal-liquid metal partitioning, more than W (e.g.,[12,13]). Variations in Os/W ratios can alo be expected at the meteorite specimen scale and this may explain the differences between ^{180}W measurements in different studies [2,3,4, this work]. Also different modal abundances of kamacite and taenite in sample cuts may contribute to any measured heterogeneities, although fractionation of Os from W between these phases appears to be minor [14].

Conclusions: We propose that a significant correlation between ε^{180}W and Os/W ratios in iron meteorites reflects radioactive alpha decay of ^{184}Os. This interpretation can explain the heterogeneity of ^{180}W in iron meteorites and can also confirm for the first time the theoretically predicted nuclear instability of ^{184}Os. Nucleosynthetic heterogeneity of ^{180}W in iron meteorites as proposed by [2] is still a viable scenario, but the magnitude of such anomalies would be limited by our observations to ca. 1ε-unit.

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