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p-Process ^{180}W anomalies in iron meteorites: Nucleosynthetic versus non-nucleosynthetic origins

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ABSTRACT

Heavy, proton-rich stable isotopes belong to the least abundant isotopes in the solar system. Their formation mechanisms and their stellar sources are most likely different from those of neutron-capture generated r- and s-process nuclei that comprise the majority of nuclides heavier than iron. Heavy p-nuclide abundances in meteorites are therefore potentially useful in deciphering distinct stellar contributions to the nascent solar system. We therefore conducted the first high-precision measurements of the heavy p-process isotope ^{180}W, achieving a typical precision of ± 0.7 ε-units for ca. 300 ng W. Measured samples comprise metals from magmatic- and non-magmatic iron meteorites, as well as metal from one H4 chondrite (NWA 926) and two reduced terrestrial basalts (from Disko Island, Greenland and the Dzheltul'ski massif from Eastern-Siberia, Russia).

The analyzed iron meteorites show clearly resolvable ^{180}W anomalies of up to $+6$ ε-units. Conversely, the chondritic metal and both terrestrial samples exhibit ^{180}W abundances indistinguishable from the standard value. As cosmogenic effects during space exposure of the meteoroids may have affected the ^{180}W budget, cosmic-ray exposure of the meteorites has to be critically evaluated. We therefore propose a method to approximate cosmogenic contributions to the ^{180}W signatures in order to unravel nucleosynthetic ^{180}W abundance anomalies. Our study reveals significant cosmogenic effects only for the longest exposed meteorites, shifting ^{180}W anomalies always to lower values (average cosmic-ray correction-factors can be estimated to lie between 0.01 and 0.30 ε^{180}W-units per 100 Myr of exposure). Cosmogenic effects for most of the analyzed meteorites therefore appear to be negligible with respect to the analytical precision achieved for iron meteorites. In addition to cosmic-ray exposure, radiogenic effects can be caused by putative decay of ^{184}Os or by decay of ^{180}Ta in its ground state. Whereas potential alpha decay of ^{184}Os could shift ^{180}W anomalies to higher values (but only up to levels that are within the analytical error of ~ 0.5 ε-units for most samples), no significant production of ^{180}W could have occurred from ^{180}Ta decay.

Notably, we identified significant and systematic abundance variations in ^{180}W between different iron meteorite groups, indicating that these isotope anomalies are characteristic for their entire parent asteroids. Our finding of decreasing excesses in ^{180}W from early formed magmatic iron meteorites ($+3.8 \pm 1.2$ ε-units) towards later formed non-magmatic iron meteorites ($+0.6 \pm 0.5$ ε-units), the analyzed chondrite and both terrestrial rocks (-0.3 ± 0.7 ε-units) may thus mirror progressive homogenization of ^{180}W in the early solar nebula. This overall trend is also supported by a co-variation between ^{180}W and metal segregation ages for the different iron meteorite groups as well as by a co-variation between ^{180}W deviations and the respective asteroidal accretion ages. Such an interpretation would suggest progressive homogenization of the solar nebula within about ~ 2.5 to ~ 6 Myr.

1. Introduction

The earliest stages in the evolution of the solar system involved a rapid, self-gravitational collapse of a dense molecular cloud core (e.g. Lada and Shu, 1990). Upon compaction, a protoplanetary disk formed, that gave birth to small asteroids and ultimately to larger planets (for recent reviews, see Chambers (2004), Nagasawa et al. (2007), Raymond (2008)). The astrophysical setting in which the earliest evolutionary steps took place is still poorly constrained. However, the heavy element inventory in the solar system had been inherited from older, long extinct stellar sources. It is likely that the formation of the presolar cloud was intimately linked to the explosion of a nearby supernova (e.g. Cameron and Truran, 1977; Jacobsen,

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2005; Meyer, 2005; Tachibana et al., 2006; Wasserburg et al., 2006). Such a scenario can not only explain nucleosynthetic abundance anomalies of some heavy stable isotopes in meteorites, but also the injection of freshly synthesized radionuclides with short half lives (<100 Ma).

One important open question in understanding the earliest history of the solar system is at which timescales isotopic mixing in the protoplanetary disk occurred. This issue may have important consequences on applying short-lived chronometers to date timescales of asteroid formation and accretion of planets. Heavy, proton-rich stable isotopes like ^{180}W are potentially useful to unravel the timescales of early solar system homogenization. Their formation mechanisms and stellar sources are most likely different from those of neutron-capture generated r- and s-process nuclei that comprise the majority of nuclides heavier than iron (for a recent review, see Arnould and Goriely (2003)). Therefore, heavy p-nuclide abundances in meteorites may also help to decipher distinct stellar contributions to the nascent solar system. However, as the relative abundances of heavy p-nuclei ($M>150$) are among the lowest of all stable isotopes ($<0.2\%$), sufficiently precise measurements of better than ±1 part per 10,000 (±1 ε-unit) are among the biggest challenges in mass spectrometry. So far, only very few lighter p-nuclides of higher abundances (^{84}Sr, ^{92}Mo, ^{120}Te, ^{130,132}Ba, ^{144}Sm, ^{146}Sm) were measured at a sufficient precision, and some of them exhibit apparent nucleosynthetic anomalies, suggesting that light p-process nuclei were heterogeneously distributed in the solar nebula (Fehr et al., 2004; Andreasen and Sharma, 2006, 2007; Carlson et al., 2007; Burkhardt et al., 2011).

Benefitting from new analytical developments in mass spectrometry we present the first high-precision data for the heavy p-process isotope ^{180}W, one of the least abundant nuclides in the solar system (0.126% relative abundance). Besides comprising an isotope nearly solely produced by the p-process, W is of additional interest as it includes the radiogenic isotope ^{182}W (decay product of now extinct ^{182}Hf; $t_{1/2}=8.9$ Myr, Vockenhuber et al., 2004) that is ideally suited to date timescales of metal segregation (e.g., Harper and Jacobsen, 1996; Kleine et al., 2002; Schoenberg et al., 2002; Yin et al., 2002). By combining ^{180}W and ^{182}W data, it is potentially possible to obtain information on the magnitude of possible nucleosynthetic ^{180}W isotope heterogeneities at a given time. We therefore employed a new analytical protocol for high precision measurements of W isotope compositions involving all five stable W isotopes and monitoring interfering Hf, Ta and Os.

Samples selected for our study include magmatic iron meteorites that formed within the first two million years of solar system history (e.g. Kleine et al., 2002; Markowski et al., 2006a; Scherstén et al., 2006; Qin et al., 2008a), non-magmatic iron meteorites and chondrites that formed within the first $\sim$10 Myr of solar system history (e.g. Kleine et al., 2008; Schulz et al., 2009, 2012) as well as terrestrial rocks which formed much later. Samples selected for this study thus cover a timespan ranging from the initial formation of the protoplanetary disk to the final consolidation of the solar system.

2. Experimental procedures

2.1. Sample preparation and chemistry

Pieces of $\sim$250 mg were cut from metal phases of all meteorites using diamond-saw blades. All metal chunks were cleaned using SiC as abrasive on a glass plate and subsequently leached in 0.05 M HNO_3 for 10 min in an ultra-sonic bath. After dissolution of up to 1 g of meteorite (in the case of Cape York up to 3 g) in aqua-regia and evaporation to dryness, samples were dried down again in concentrated HNO_3 to remove any possible organic interferences. After drying-down, the samples were redissolved in 6 N HCl/0.2 N HF. Tungsten was separated by anion exchange using BioRad AG 1X8 resin. The procedure was modified from methods previously published by Kleine et al. (2004) and Schulz et al. (2009) to achieve a better purification from interfering Hf. This was achieved by doubling the amount of 9 M HCl/0.01 M HF in the rinsing step of the 2nd stage (clean-up) chemistry and by using 6 M HNO_3/0.2 M HF instead of 7 M HCl/1 M HF in the W elution step. In some cases, one meteorite was split and processed on two columns (each containing 4.5 ml of resin) at the same time and the W cuts were later combined. This prevented overloading of the ion exchange column and improved column yields. Tungsten yields were typically in the range of $\sim$60–80 %. Total procedural blanks for W were $\sim$300 pg.

2.2. Mass spectrometry

2.2.1. Instrumentation and measurement protocols

Analyses were performed in the clean lab facilities at Köln/Bonn using a Thermo Finnigan^{TM} Neptune Multicollector ICPMS, equipped with two 10^{-12} Ω amplifiers. The instrument is also equipped with a Pfeiffer on Tool Booster^{TM} interface pump, permitting measurements with Jet sample cones of a large aperture. These interface cones can significantly increase instrumental sensitivity, but may create anomalous mass fractionation effects as discussed below. The W isotope measurements were performed in static mode by simultaneous collection of the ion beams of masses 178 (^{178}Hf), 180 (^{180}W, ^{180}Hf, ^{180}Ta), 181 (^{181}Ta), 182 (^{182}W), 183 (^{183}W), 184 (^{184}W, ^{184}Os), 186 (^{186}W, ^{186}Os) and 188 (^{188}Os) with Faraday cups. Instrumental mass bias was corrected relative to three different isotope pairs in order to check for analytical artifacts or nucleosynthetic effects. Measured values for ^{180}W/^{183}W, ^{182}W/^{183}W and ^{184}W/^{183}W were exponentially corrected for instrumental mass bias relative to ^{186}W/^{183}W, using a ratio of 1.98594. Furthermore, ^{180}W/^{184}W, ^{182}W/^{184}W and ^{183}W/^{184}W were corrected relative to ^{186}W/^{184}W using a ratio of 0.92767 and ^{180}W/^{184}W, ^{182}W/^{184}W as well as ^{186}W/^{184}W relative to ^{183}W/^{184}W using a ratio of 0.46712 (values from Völkening et al. (1991)). In all cases, samples were bracketed by standard measurements and are reported in ε^{18i}W, which is the deviation of the measured ratios from the terrestrial AMES standard value in parts per 10,000). If not otherwise stated, all ε^{180}W and ε^{182}W values mentioned in the text represent the ratios ^{180}W/^{183}W and ^{182}W/^{183}W normalized to ^{186}W/^{183}W.

Tungsten isotope compositions were typically measured at signal intensities of 10–40 V for ^{184}W, corresponding to $\sim$40–160 mV on ^{180}W, as typically obtained for 100–400 ng of W using X skimmer cones and 50–200 ng using Jet sampler cones in low resolution mode. Internal errors on measured ^{180}W abundances obtained for such signal intensities were typically as good as ±70 ppm (2σ) and the reproducibility of standard measurements conducted in the same measurement session was typically ±100 ppm for ^{180}W, ±14 ppm for ^{182}W, ±14 ppm for ^{183}W, ±12 ppm for ^{184}W and ±25 ppm for ^{186}W (2σ). A long-term reproducibility was additionally evaluated over a period of 1 yr in 9 different measurement sessions, each comprising up to 20 individual standard runs (Fig. 1). The long-term reproducibility for ^{180}W was calculated to be ±172 ppm (2σ). For the last 4 measurement sessions the reproducibility improves to about ±100 ppm, nearly identical to the precision achievable on a single day. All of the nine measurement sessions were carried out using low-resolution mode and X-skimmer cones. The short-term reproducibility using Jet cones is comparable to the X cone set up in all resolution modes. However, the long-term reproducibility for six measurements using both cone set-ups and varying resolution modes yielded a value of ±272 ppm, significantly higher compared to the low resolution/X-cone set-up (Fig. 1).