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Excess ^{180}W in IIAB iron meteorites: Identification of cosmogenic, radiogenic, and nucleosynthetic components

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ABSTRACT

The origin of ^{180}W excesses in iron meteorites has been a recently debated topic. Here, a suite of IIAB iron meteorites was studied in order to accurately determine the contribution from galactic cosmic rays (GCR) and from potential decay of ^{184}Os to measured excesses in the minor isotope ^{180}W. In addition to W isotopes, trace element concentrations (Re, Os, Ir, Pt, W) were determined on the same samples, as well as their cosmic ray exposure ages, using ^{36}Cl–^{36}Ar systematics. These data were used in combination with an improved model of GCR effects on W isotopes to correct effects resulting from neutron capture and spallation reactions. After these corrections, the residual ^{180}W excesses correlate with Os/W ratios and indicate a clear contribution from ^{184}Os decay. A newly derived decay constant is equivalent to a half-life for ^{184}Os of $(3.38 \pm 2.13) \times 10^{13}$ a. Furthermore, when the data are plotted on an Os–W isochron diagram, the intercept ($\varepsilon^{180}\text{W}_\text{i} = 0.63 \pm 0.35$) reveals that the IIAB parent body was characterized by a small initial nucleosynthetic excess in ^{180}W upon which radiogenic and GCR effects were superimposed. This is the first cogent evidence for *p*-process variability in W isotopes in early Solar System material.

1. Introduction

The nebular material (gas and dust) from which the Solar System formed was contributed by multiple types of stellar sources (e.g., red giants, supernovae). Measuring the isotopic composition of various Solar System objects can help to determine the degree of mixing of the starting materials, which can elucidate the physical process that operated in the protoplanetary disk. In addition, isotopic studies may help to identify specific stellar sources that contributed material to the solar nebula, and perhaps, the birth environment of the Solar System (e.g., Boss, 2017; Cameron and Truran, 1977; Dwarkadas et al., 2017). Isotopes produced by the *p*-process are among the rarest in the Solar System (e.g., Arnould and Goriely, 2003; Rauscher et al., 2013). As such, *p*-isotopes could provide a sensitive tracer of mixing processes in the solar nebula. In particular, the rare isotope ^{180}W (relative abundance 0.12%; Völkening et al., 1991a) may provide a powerful tool to constrain the degree of mixing and/or stellar sources that fed the solar nebula.

Compared to terrestrial material, excesses of ^{180}W occur in members from many groups of magmatic iron meteorites, but their origin is debated. Excess ^{180}W may represent *p*-process heterogeneity in the solar nebula (Schulz et al., 2013), radiogenic ingrowth on the parent body from ^{184}Os decay (Peters et al., 2014), or spallogenic production due to galactic cosmic ray (GCR) interactions while the material was in outer space (Cook et al., 2014).

Effects from GCRs may increase or decrease ^{180}W concentrations due to the complex interplay between neutron capture and spallation reactions. The final induced effect depends on several variables, including sample depth below the surface, elemental composition, and exposure duration (e.g., Cook et al., 2014; Leya and Masarik, 2013). Without an accurate correction for GCR effects, the true isotopic composition cannot be determined.

Correlations between ^{180}W and Os/W ratios suggest that decay from ^{184}Os may be a contributing factor to ^{180}W excesses (Cook et al., 2014; Peters et al., 2014). However, the W isotopic data of Peters et al. (2014) lacked corrections for GCR effects, and in some cases, also for nucleosynthetic variations on effects on other W isotopes that can cause apparent ^{180}W excesses (e.g., in IVB irons). In Cook et al. (2014), corrections were made for both effects, yet many samples had to be omitted from the resulting isochron because the required parameters needed to properly correct for

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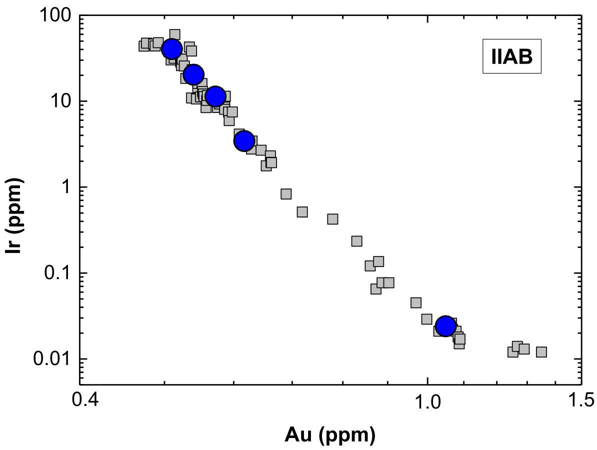

Fig. 1. Concentrations of Ir versus Au in IIAB iron meteorites. The five samples included in this study (Sikhote-Alin, North Chile, Braunau, Cincinnati, Forsyth County) are shown by the blue circles. Data from Wasson et al. (2007). (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)

GCR effects were unknown. Furthermore, in both studies, isochrons were constructed using mixtures of irons from several magmatic groups, which originated on separate parent bodies and with different crystallization histories and ages. The subsequent values for the ^{184}Os decay constant (λ^{184}Os) determined in these two studies differ by a factor of ≈2. Hence, the need exists to better constrain the λ^{184}Os value and to properly determine the magnitude of any ^{184}Os decay related to observed ^{180}W excesses. In addition, the application of ^{184}Os–^{180}W systematics may be a useful approach to determine the initial ε^{180}W in Solar System materials.

A suite of five group IIAB irons were chosen to decipher the exact nature of ^{180}W excesses. Group IIAB irons have several characteristics that make them ideally suited for this: 1) their formation via extensive fractional crystallization (Scott, 1972; Wasson et al., 2007) permits the sampling of a wide range in Os/W ratios required to construct an isochron; 2) their metal–silicate segregation and crystallization ages are independently known from ^{182}Hf–^{182}W and ^{187}Re–^{187}Os systematics, respectively (Cook et al., 2004; Kruijer et al., 2014); 3) their cosmic ray exposure (CRE) ages tend to be shorter than the CRE ages of other magmatic groups (e.g., Herzog, 2004), minimizing potential GCR effects; and 4) they lack a resolvable deficit in *s*-process W isotopes that is observed in some other iron groups (Cook et al., 2014; Kruijer et al., 2014; Qin et al., 2008), eliminating the need to correct for collateral *s*-deficit effects on ^{180}W. Here, an isochron based on GCR-corrected W isotopic ratios is used to re-assess the λ^{184}Os value and to determine the initial ^{180}W/^{184}W ratio of the IIAB iron meteorite parent body.

2. Methods

2.1. Samples and preparation

The five IIAB irons were chosen to sample widely different portions of the crystallization sequence for this group (Fig. 1). Separate metal pieces were cut from the same sample mass for the various analytical objectives required for each sample; adjacent pieces were used for the analyses of W isotopes and for siderophile trace element concentrations. Samples were cut with a slow-speed saw, polished with SiC paper, and rinsed with ethanol. Pieces of ≈500 mg were prepared for W isotopic analyses; two pieces (≈100 mg each) were cut for analyses of radionuclides (e.g., ^{36}Cl, ^{41}Ca) and noble gases (e.g., Ar), respectively; pieces ranging from ≈330 to ≈460 mg were prepared for trace element determinations. In the case of Braunau, Cincinnati, and Forsyth County, the same iron meteorite chunks were sampled as in the previous study by Cook et al. (2014). For Forsyth County, insufficient material remained to make a new analysis of W isotopes; thus, the W isotopic data from Cook et al. (2014) are used here.

2.2. Tungsten separation and isotopic analyses

The separation of W from the matrix and the subsequent isotopic analyses follow the methods in Cook and Schönbächler (2016) and are briefly summarized here. Samples were digested in 15 ml of concentrated HNO_3:HCl (2:1) at 130 °C. They were dried, fluxed in concentrated HCl, dried again, and dissolved in 0.5 M HCl–0.5 M HF. Tungsten was separated using a two-step anion exchange column procedure using HCl–HF media. Purified W was dried in a mixture of HNO_3:H_2O_2:HF (1:1:0.5) and then dissolved in 0.5 M HNO_3–0.2 M HF for isotopic analysis. Isotopic measurements were made using a Neptune Plus MC-ICPMS at ETH-Zürich. All samples, meteoritic and terrestrial, were measured once, bracketed by measurements of the NIST SRM 3163 W solution. Instrumental mass bias was corrected with the exponential law with either $^{186}\text{W}/^{183}\text{W} = 1.98594$ or $^{186}\text{W}/^{184}\text{W} = 0.927672$ (Völkening et al., 1991a). All measurements were corrected for nuclear field shift (NFS) effects (Cook and Schönbächler, 2016). Five replicates of the NIST Fe–Ni steel SRM 129c were chemically processed and analyzed during the same session as the IIAB irons to validate the accuracy of the measurements; this material also served as an external standard to define the long-term reproducibility of the entire analytical method. The external reproducibility (2SD), based on a total of 20 replicate analyses of the NIST Fe–Ni steel SRM 129c, on ε^{i}W values are ±0.05 for ε^{184}W (6/3), ±0.49 for ε^{180}W (6/4), ±0.08 and for ε^{182}W (6/4) (see Cook and Schönbächler, 2016; Table 2). The notation "(6/3)" and "(6/4)" indicates the ratio used for the mass bias correction (i.e., ^{186}W/^{183}W or ^{186}W/^{184}W).

2.3. Trace element analyses

Trace element concentrations (Re, Os, Ir, Pt, W) were determined at the University of Maryland (College Park) by isotope dilution. Separation of Re, Os, Ir, and Pt followed the methods in Walker et al. (2008). Samples were digested with 5 ml of concentrated HNO_3 and 2.5 ml of concentrated HCl in Pyrex® Carius tubes and equilibrated with a combined platinum-group element spike (^{191}Ir and ^{194}Pt) and a Re–Os spike (^{185}Re and ^{190}Os) for ≥24 h at 220 °C (Shirey and Walker, 1995). Osmium was separated by solvent-extraction (Cohen and Waters, 1996), further purified using micro-distillation (Birck et al., 1997), and then analyzed by N-TIMS (ThermoScientific Triton). A natural Os in-house standard was also measured to correct for instrumental mass fractionation, which was negligible for these measurements (≪0.1%). The remaining Re, Ir and Pt were purified using an anion exchange column procedure (Rehkämper et al., 1997). Purified and separated Re, Ir, and Pt were evaporated to dryness and dissolved in 0.8 M HNO_3 for analysis by MC-ICPMS (ThermoScientific Neptune Plus), using natural Re, Ir, and Pt standards to correct for instrumental mass fractionation. The separation of W is based on the methods from Touboul and Walker (2012). Samples were digested in 5 ml of 8 M HCl and equilibrated with a ^{182}W spike at 130 °C. Tungsten was purified using an anion exchange column and dissolved in 0.8 M HNO_3 for analysis by ICPMS, using a natural W standard to correct for instrumental mass fractionation. The total blanks for Os, Re, Ir, Pt, and W were 1, 0.4, 0.5, 4, and 30 pg, respectively, which are negligible for all samples. The uncertainties (2SE) on Os, Re, Ir, Pt, and W concentrations are 0.1%, 0.1–0.2%, 0.1–0.2%, 0.1–0.2%, and 0.1%, respectively.