TUNGSTEN ISOTOPIC COMPOSITION CORRECTED FOR COSMIC RAY EFFECTS AND THE Hf-W AGE OF IRON METEORITES

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Abstract: We report a method to correct W isotopic compositions in iron meteorites for cosmic-ray-induced variations [1]. This allows us to deduce that at least some magmatic irons segregated within less than 1 Myr of CAI formation [2].

Introduction: Magmatic iron meteorites are believed to sample the metal cores of planetesimals that formed by metal–silicate segregation followed by fractional crystallization of the metallic melt [3]. Core formation in planetesimals can in principle be dated using the ^{182}Hf-^{182}W chronometer applied to iron meteorites [2, 4-9]. However, production and burnout of W isotopes due to a long exposure of the sample to cosmic rays and interaction with thermal neutrons can lower the ^{182}W/^{184}W ratio in iron meteorites, resulting in apparently older ages [10-11]. Cosmogenic interactions affect all W isotopes, depending on the relevant cross-section and relative abundance of the isotope. Furthermore the effect depends on meteorite size, sample depth and exposure age. Tungsten isotope variations have been reported in different iron meteorites [e.g. 2, 6-9], but their cause remains debated. In theory, differences in ^{182}W/^{184}W ratio are expected if iron meteorites segregated at different times or if accretion and differentiation were protracted. However, isotopic heterogeneities may also be due to nucleosynthetic anomalies or cosmogenic effects. It is therefore important to understand and constrain the different processes that can modify the W isotopic composition of meteorites.

We present here the first experimental evidence for cosmic ray isotopic effects on W, with a study of samples taken from slabs from two large iron meteorites, Carbo and Grant, for more detail see [1]. The cosmogenic noble gas concentrations across these slabs had previously been measured and hence the preatmospheric center of the meteorites is roughly known [e.g. 12, 13]. Based on new nuclear physics parameters, we developed a physical model which allows us to correct for the cosmogenic effect on W isotopes provided that the ^{3}He concentration of the sample and an independent estimate of the exposure age are known.

Nucleosynthetic effects: Tungsten isotopes 182, 183, 184 and 186 are produced by *s*-and *r*-processes, whereas ^{180}W is produced by the *p*-process [14-15]. Nucleosynthetic anomalies in iron meteorites, have been reported for several elements [16-18]. However, all ^{184}W/^{183}W ratios measured in this study are equal to the terrestrial standard within uncertainty. We can therefore assume that no nucleosynthetic effect on W due to *s*-and/or *r*-processes could be measured at our level of precision. Possible anomalies on ^{180}W would not be indicative for any *s*-and/or *r*-nucleosynthetic contributions. Furthermore, the precision on ^{180}W obtained up to now is not sufficient to detect a possible nucleosynthetic effect.

Results: In Figure 1, we present the first evidence of variable W isotopic composition in a single meteorite as a function of depth.

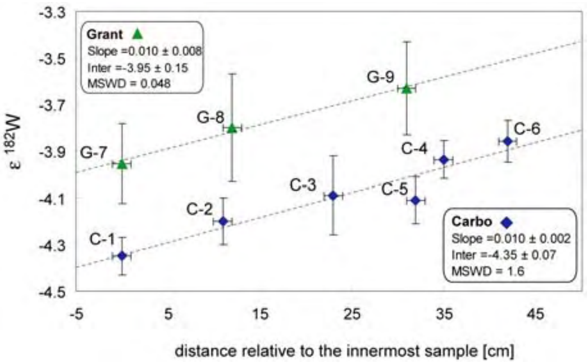

Fig. 1: Correlation between the ε^{182}W value and the distance relative to the sample closest to the preatmospheric centre. For exact sample location see [1]. $\varepsilon\,^{182}$W is the deviation from the terrestrial standard value in parts per 10,000.

All W isotopic measurements were performed using a Nu Plasma MC-ICPMS at ETH Zürich. Each sample has been measured up to 18 times in different sessions. The data were corrected for mass bias using ^{186}W/^{183}W = 1.985936 [19]. Along two profiles in Carbo (samples C1-C4 and C5-C6), ε^{182}W increases by ~ 0.5ε units from the preatmospheric centre towards the preatmospheric surface (Fig. 1). The same trend is observed for Grant (~0.3ε units, samples G7-9). Figure 1 represents the first experimental evidence for the modification of W isotopic composition by galactic cosmic rays, because the effect varies with shielding [10-11]. Figure 2 shows ε^{182}W versus the ^{3}He concentrations of the samples as found in the literature. Note that ^{3}He and cosmogenic W are produced by two different processes; while ^{3}He is produced by high energy particles mainly from Fe and Ni, burnout and production of W isotopes mainly occurs by slow neutrons, the flux of which increases with shielding. Thus ε^{182}W values inversely correlate with the ^{3}He concentration.

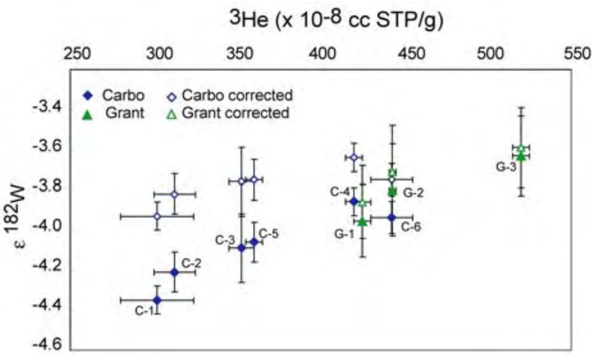

Fig. 2: ε^{182}W versus ^{3}He concentration taken from published data. Solid symbols represent the measured ratios without cosmogenic correction, whereas the open symbols are corrected data as explained in the text. Error bars on all ratios are the 2σ error on the measurements combined with the error on the bracketing standards. The uncertainty of the model calculations is estimated to be about 50%.

Model: We modeled the modification of the W isotope composition due to cosmic ray interactions as a function of the ^{3}He concentration and the exposure age for iron meteorites with radii between 5 and 85 cm (Fig. 3). We found that ^{3}He concentrations inversely correlate with the effect on the ε^{182}W [1].

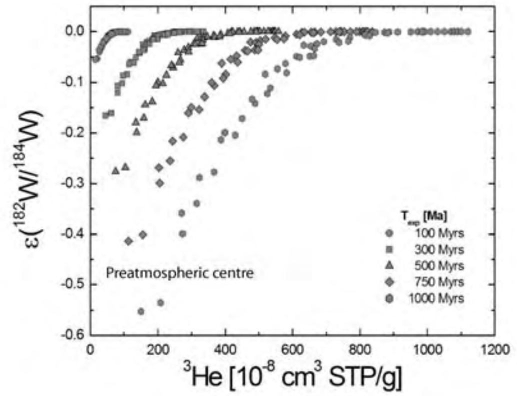

Fig.3: Model of the cosmogenic effect on ε^{182}W versus [^{3}He], for meteorites having different exposure ages and radii of 5 - 85cm.

Cosmic-ray effects were calculated by integrating depth- and size dependent spectra of primary and secondary particles with the excitation functions of the relevant nuclear reactions. Particle spectra were derived by following the trajectories of primary GCR-particles and calculating production and transport of secondary particles using Monte-Carlo techniques. Excitation functions for (n,γ)- and (n,2n)-reactions and production cross sections of ^{3}He from Fe and Ni are given in [20, 21].

In this model, samples close to the pre-atmospheric centre have a low ^{3}He concentration and the largest effect on ε^{182}W. The largest modelled meteorite here (r=85cm) also shows the largest effect, but for meteorites with even larger radii the cosmogenic effect must inverse, as had e. g. been shown by Masarik for Toluca [10]. As a consequence, curves increase very likely towards zero at very low ^{3}He concentrations for very large meteorites.

Similar calculations have also been done for the ^{186}W/^{183}W ratio, the ratio used for the mass bias correction. The overall corrections on the ε^{182}W also consider this latter correction as shown in Figure 2. We assumed an exposure age of 850 ±140 Ma for Carbo and 695±65 Ma for Grant [22]. The correction lies between 0.42ε and 0.22ε units for Carbo and between 0.09ε and 0.04ε units for Grant. We consider the sample with the smallest correction as the best estimate for the true W isotopic composition of Carbo and Grant, which corresponds to ε^{182}W =–3.64±0.11 and ε^{182}W =–3.59±0.11 respectively. Finally, we applied this method to other iron meteorites for which we measured [^{3}He] and found that the real ^{182}W deficits, are similar to those measured for Allende CAIs [2]. This indicates that at least some iron meteorites formed within 0.0±1.0 Myr of the start of the solar system.

References: [1] Markowski et al. *(submitted to EPSL)* [2] Kleine et al. *GCA 69, 5805-5818.* [3] Scott and Wasson (1975) *Rev. Geophys. Space Phys. 13,* 526–546. [4] Lee and Halliday (1995) *Nature 378,* 771-774. [5] Harper and Jacobsen (1996) *GCA 60,* 1131-1153 [6] Horan et al (1998) *GCA 62*, 545-554. [7] Lee (2005) *EPSL 237*, 21-32. [8] Schérsten et al. *EPSL (in press)* [9] Markowski et al. *EPSL (in press).* [10] Masarik (1997) *EPSL 151*, 181-185. [11] Leya et al. (2003) *GCA 67*, 529-541. [12] Signer and Nier (1960) *J. Gephy. Res. 65,* 2947-2964 [13] Ammon and Leya, (2006) LPSC XXXVII [14] Anders and Grevesse (1989) *GCA 53*, 197-214. [15] Norman and Schramm (1983) *Nature 304*, 515-517. [16] Quitté et al. (2006) *EPSL (in press)* [17] Dauphas et al. (2004) *EPSL 226*, 465-475 [18] Chen et al. (2003) *LPI XXXIV* #1789 [19] Völkening et al. (1991) *Int. J. Mass Spectr. Ion Processes 107, 361-368* [20] Kinsey R. (1979) ENDF-102 *Data formats and Procedured for Evaluated Nuclear Data File ENDF* [21] Leya et al. (2004) *MAPS 39*, 367-386 [22] Voshage and Feldmann (1979) *EPSL 45*, 293-308