

COSMOGENIC STABLE ISOTOPE EFFECTS IN CARBO. L. Qin¹ and N. Dauphas^{2,3}. ¹Department of Terrestrial Magnetism, Carnegie Institution of Washington, 5241 Broad Branch Road, NW, Washington, DC 20015, USA. ²Origins Laboratory, Department of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, Chicago IL 60637, USA. ³California Institute of Technology, Division of Geological & Planetary Sciences. (E-mail: lqin@ciw.edu)

Introduction: The ^{182}Hf - ^{182}W decay system ($t_{1/2}=9$ My) is useful to date early Solar System processes, notably metal-silicate differentiation [1-10]. Hf-W systematics of refractory inclusions indicate that these objects formed with an initial $^{182}\text{Hf}/^{180}\text{Hf}$ ratio of $\sim 1 \times 10^{-4}$ and $\epsilon^{182}\text{W}$ of -3.47 ± 0.20 [6]. Recent high precision studies revealed negative $\epsilon^{182}\text{W}$ values relative to CAIs for some iron meteorites [3, 5-10]. Taken at face value, this would indicate that core formation in the parent bodies of these meteorites predated CAI formation. Markowski et al. [9] measured the W isotopic compositions along two depth profiles in the Grant (IIB) and Carbo (IID) iron meteorites. They showed that these iron meteorites were affected by cosmogenic effects corresponding to a decrease of $\epsilon^{182}\text{W}$ by capture on W isotopes of secondary thermal neutrons. In a previous study, we analyzed pieces of Grant from various locations and found a similar trend [10]. However, the measurements were not very precise due to the low W concentration of this meteorite. One major challenge in establishing the ^{182}Hf - ^{182}W system as a reliable chronometer is to find a way to accurately correct for these cosmogenic effects [11, 12]. Knowledge of exposure ages alone is not sufficient because the cosmic irradiation is also modulated by the depth of burial in the parent-body. A robust way of estimating cosmic-ray effects would be to identify correlated cosmogenic effects in other isotopes which are also produced by neutron capture processes. A recent study found Os isotopic variations in IVB meteorites [13]. The correlation between the various Os isotopes is consistent with model predictions of neutron-capture. The magnitude of the cosmogenic effect is largest in Tlacotepec because of its long exposure age. Thus, Os isotope compositions can potentially be used to monitor neutron capture effects in iron meteorites. In this study we revisit W isotope variations in Carbo.

Methods: *Chemical separation.* We recently developed an ion exchange procedure coupling one cation-exchange column and several anion-exchange columns to remove matrix and interfering elements in iron meteorites [14,15]. The same column chemistry was used in this study. The final W-cut was evaporated to dryness in aqua regia more than 5 times, followed by several drying down in mixtures of HNO_3 and H_2O_2 . This was done to remove Os and organic matter from the resin. HClO_4 was used in our previous work,

but was avoided in this study. We found that it can potentially cause unwanted W isotopic fractionation in the Neptune MC-ICPMS, which was not observed on the Isoprobe MC-ICPMS. The Os/W ratios in the final solutions were lower than 10^{-4} .

Mass spectrometry. Tungsten isotopic analyses were performed with a Thermo Scientific Neptune HR-MC-ICPMS. The instrument is equipped with 9 faraday cups. The sample solutions were introduced into the mass spectrometer using an Apex desolvating nebulizer. Measurements were performed in low-resolution mode. Ion intensities of ^{179}Hf , $^{180}\text{W}+^{180}\text{Hf}+^{180}\text{Ta}$, ^{181}Ta , ^{182}W , ^{183}W , $^{184}\text{W}+^{184}\text{Os}$, $^{186}\text{W}+^{186}\text{Os}$ and ^{188}Os were measured on Faraday collectors L4, L3, L2, L1, Center, H1, H2, H3. Measuring ^{179}Hf , ^{181}Ta and ^{188}Os allowed us to monitor and correct isobaric interference from Hf, Ta and Os. The samples were run at 30-50 ppb concentration. Measured intensities were in the range of 4-6 V on ^{186}W . The measurements were done in sequences of 20 cycles, with each cycle integrating ion intensities for 8 s. The sample measurements were bracketed by standard measurements to control the drift in instrumental mass fractionation with time. Sample-standard bracketing was repeated 10 times for each sample. The W concentration in the sample solution was strictly matched with that of the bracketing standard to within ± 2 -3%. The NIST 3163 W standard was used as the reference material. Measured $^{182}\text{W}/^{183}\text{W}$ and $^{184}\text{W}/^{183}\text{W}$ ratios were corrected for mass fractionation by normalizing to a fixed $^{183}\text{W}/^{186}\text{W}$ ratio of 0.501800 using the exponential law.

Results: More than 15 pieces of Carbo were analyzed. Three pieces from adjacent locations were also studied. They showed identical $\epsilon^{182}\text{W}$ of -4.13 ± 0.14 , -4.07 ± 0.11 , -4.14 ± 0.08 , and identical $\epsilon^{184}\text{W}$ values of -0.03 ± 0.05 , -0.02 ± 0.07 and -0.03 ± 0.06 . $\epsilon^{184}\text{W}$ is useful to monitor accuracy of the measurements. The average $\epsilon^{182}\text{W}$ value (-4.11 ± 0.05) of the three pieces is identical to our previous published value for Carbo of -4.09 ± 0.08 measured on an Isoprobe MC-ICPMS.

Another 10 pieces of Carbo came from a slab and their locations and noble gas concentrations are well documented, which makes these samples ideal for studying cosmogenic effects. The $\epsilon^{182}\text{W}$ values in these samples vary from -3.9 to -4.3, a range similar to that obtained by [9]. We analyzed meteorite pieces from

locations adjacent to those sampled by [9] and there is perfect agreement between the 2 studies. The variations in $\epsilon^{182}\text{W}$ correlate well with ^3He concentrations (also produced by interaction with cosmic rays) and distance from pre-atmospheric center (Fig. 1, 2). The samples farthest away from the pre-atmospheric center have the least negative $\epsilon^{182}\text{W}$ values and the highest ^3He concentrations (Fig. 1, 2). The measurements also agree with model predictions of cosmogenic effects based on the JEF and KASKAD libraries (Fig. 2 and ref [10]). The least negative $\epsilon^{182}\text{W}$ value among these samples is ~ -3.90 , a value much more negative than the pre-exposure values estimated for iron meteorites from other groups. Model predictions suggest that the W isotopic compositions of samples at the pre-atmospheric surface should not be affected by cosmogenic effect. The lack of more positive values indicates that samples closest to the pre-exposure surface are probably missing.

Most $\epsilon^{184}\text{W}$ values are identical with the standard within error. Some values are slightly negative but the effects are barely resolvable outside 2σ error.

Conclusions: The W isotopic analyses on a Thermo Scientific Neptune HR-MC-ICPMS reported in this study agree with those performed on an IsoProbe MC-ICPMS. This study confirms the conclusion of [9] that $\epsilon^{182}\text{W}$ correlates with ^3He concentrations in Carbo. We plan to expand this study to other elements to devise methods for identifying and correcting cosmogenic effects in iron meteorites.

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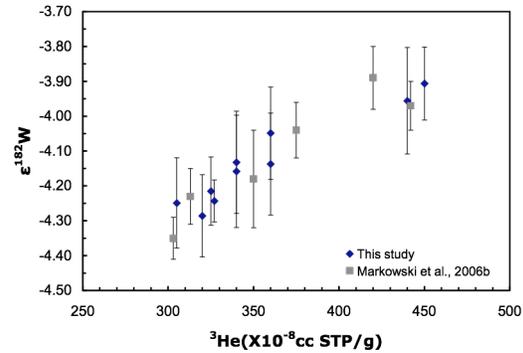


Fig. 1. W isotopic compositions of Carbo versus ^3He .

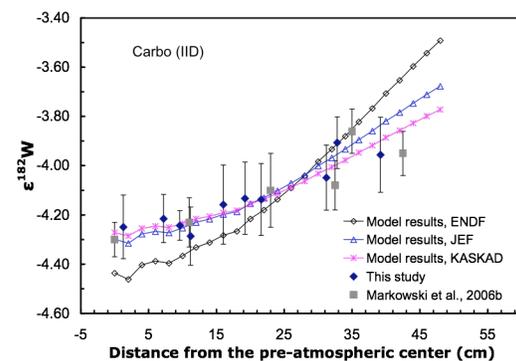


Fig. 2. W isotopic compositions of Carbo versus the distance from the pre-atmospheric center.