EVIDENCE FOR RHENIUM ISOTOPIC FRACTIONATION DURING CRYSTALLIZATION AND MIXING IN THE IIAB IRON CORE. Q. -F. Mei[1], S. Sharma[1], I. Leya[2], M. Humayun[1], [1]National High Magnetic Field Laboratory and Department of Earth, Ocean & Atmospheric Science, Florida State University, Tallahassee, FL 32310, USA (qfmei@magnet.fsu.edu), [2]Physical Institute, Space Science and Planetology, University of Bern, Sidlerstrasse 5, 3012 Bern, Switzerland.

Introduction: The short-lived ^{182}Hf-^{182}W system has been widely used to constrain the timescale of metal-silicate separation (e.g., core formation in asteroids) [1-5]. High precision W isotope measurements (±3-5 ppm) could achieve a time resolution of 0.3-0.5 Ma [6-7]. However, applications of this chronometer to iron meteorites are hampered by neutron capture reactions on W and Re isotopes that alter the W isotope compositions [8-10], only partially corrected by proxy neutron dosimeters, e.g., Pt [11-12].

Neutron capture on ^{184}W and ^{186}W yields isotopes of Re (^{185}Re and ^{187}Re). An initial goal of this study was to directly measure the neutron capture rate on W isotopes used for normalization purposes (^{184}W, ^{186}W) in the IIAB iron meteorites since galactic cosmic ray (GCR) irradiation ages are highest for the most evolved irons (high W/Re) and pre-irradiation W and Re isotope compositions could be obtained from the least evolved and irradiated irons.

During fractional crystallization, some elements exhibit intrinsic mass dependent fractionation, e.g., Ru isotopes [13]. Previous Re isotopic analyses on IVB irons did not find any systematic changes across the fractional crystallization spectrum, but noted Re isotopic differences in IIABs between Negrillos and Coahuila [14]. The variable δ^{187}Re in IIAB iron meteorites [14] was suspected to result from GCR neutron irradiation, an effect that would be more prominent at higher W/Re ratio. The IIAB iron meteorites have one of the largest ranges of W/Re ratios (from 0.55 to 523; [15]) in a single group of iron meteorites and have a substantial range of GCR exposure ages making IIABs an ideal group within which to investigate the W-Re neutron capture network.

Here, we report combined W-Re-Pt isotopic data for IIAB iron meteorites to examine intrinsic and cosmogenic isotope effects over the broadest compositional range observed in a single iron meteorite group.

Analytical Methods: Aliquots of 17 IIAB iron meteorites with W/Re ratios ranging from 0.68 to 258 were digested in aqua regia, and the W, Re, and Pt fractions were purified with a two-step ion-exchange chromatography procedure modified from previous studies [11, 14, 16].

High precision W and Re isotopic measurements were performed on a Thermo Neptune™ MC-ICP-MS following previous studies [7, 14]. High precision W isotope data were obtained by internal normalization, and instrumental bias for Re isotopes was corrected by the W-doping method [14]. All of the W and Re isotopic ratios were normalized to ^{186}W/^{184}W = 0.92767 using the exponential law. The ^{185}Re/^{184}W ratios in the spiked Re solutions were maintained in the range of 0.60 to 0.75 to minimize the effects of WH^+ and ReH^+ [14]. The isobaric Os interference on Re was removed by sparging with Ar in nitric acid prior to analysis. The ^{190}Os peak was monitored to correct for any residual ^{187}Os interference. The ^{187}Os/^{185}Re correction applied to samples was generally lower than 0.05‰.

The W isotopic composition is reported as ε^{18i}W: $\varepsilon^{18i}W = [(^{18i}W/^{184}W_{sample})/(^{18i}W/^{184}W_{NIST\ SRM\ 3163})-1]\times10000$, where i refers to 2 or 3. The Re isotopic composition is reported as δ^{187}Re: $\delta^{187}Re = [(^{187}Re/^{185}Re_{sample})/(^{187}Re/^{185}Re_{BDH})-1]\times1000$. BDH™ Re solution and NIST SRM 3163 W solution were used as isotopic reference standards.

Results: The W and Re isotopic compositions of the analyzed IIAB samples are shown in Fig. 1.

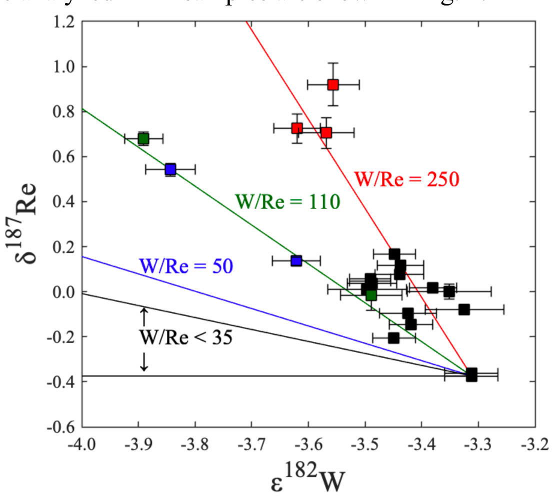

Fig. 1. Plot of δ^{187}Re (‰) versus ε^{182}W for IIAB iron meteorites. Error bars represent 2SE. Lines represent model variations of δ^{187}Re and ε^{182}W by GCR neutron capture for irons color-coded by W/Re ratio [17].

Negrillos has the lowest δ^{187}Re value (-0.37 ± 0.05‰) and highest ε^{182}W value (-3.31 ± 0.05) in the measured IIAB samples, and has the lowest GCR exposure age of all studied IIABs, < 50 Ma [18]. Agoudal shows the highest δ^{187}Re value (0.92 ± 0.09‰). Mount Joy has an ε^{182}W value of -3.84 ± 0.04 and a δ^{187}Re value of 0.54 ± 0.03‰, and Sandia Mountains was reported in [19]. A systematic increase in δ^{187}Re occurs in the IIAB fractionation sequence (Fig. 2).

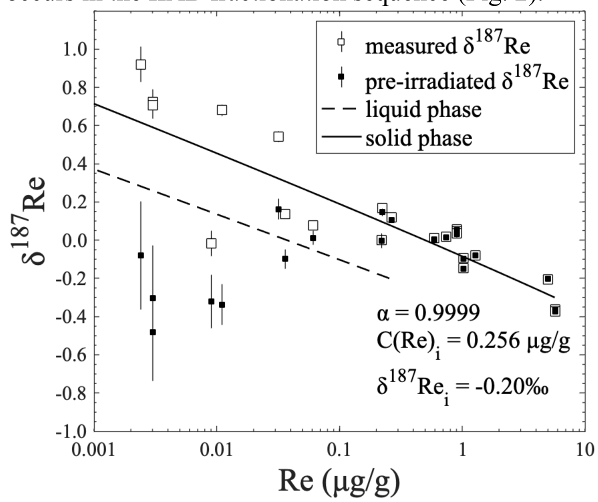

Fig. 2. Plot of measured (open squares) and cosmogenic effect-corrected (filled squares) Re isotope compositions of IIAB iron meteorites *vs*. Re concentrations [20]. Error bars represent 2SE. The model curves show Rayleigh fractionation of Re isotopes with α = 0.9999 for solids (solid line) and residual liquid (dashed line) with initial sulfur of 18% [21].

Discussion: The presence of intrinsic mass dependent Re isotope fractionation in the IIABs nullified the original goal, but provided new insights on the complexities of core solidification in IIABs. Neutron capture on ^{186}W creates the correlated increase in δ^{187}Re with ε^{182}W (Fig. 1). A modified version of the model of Leya and Masarik [17] was employed to correct the GCR effects on Re isotopes including the effects of capture on W, Re, Os, and Pt isotopes on δ^{187}Re. Meteorites in the IIAB group are assumed to have a uniform pre-irradiated ε^{182}W represented by the least irradiated members (i.e., Negrillos) [22]. The model correlation between the change in ε^{182}W and the change in δ^{187}Re [17] relative to Negrillos was used to restore the original Re isotope composition. A plot of measured and neutron capture-corrected δ^{187}Re of IIAB iron meteorites with Re concentrations are shown in Fig. 2.

The pre-irradiated δ^{187}Re values in the IIAB group are neither constant, nor monotonic with decreasing Re contents (Fig. 2). The pre-irradiated δ^{187}Re increases from the high Re end to Re ~ 0.1 μg/g, which is attributed to Rayleigh fractionation of a metallic liquid [21]. A reversal in the pre-irradiated δ^{187}Re towards lower Re concentrations (high W/Re ratios) is observed. The reversal in the δ^{187}Re values of IIAB group is accompanied by a reversal in the Re/Os ratios *vs*. Re content (Fig. 3).

Increasing Re/Os and δ^{187}Re with decreasing Re content is predicted by closed system fractional crystallization. The reversals of the Re/Os ratios (red curve, Fig. 3) and δ^{187}Re in the middle of the IIAB sequence imply a mixing of a primary liquid that drips into the core from an actively accreting asteroidal mantle. The new δ^{187}Re data indicate that fractionation in the IIAB core is more complex than closed system Rayleigh fractionation.

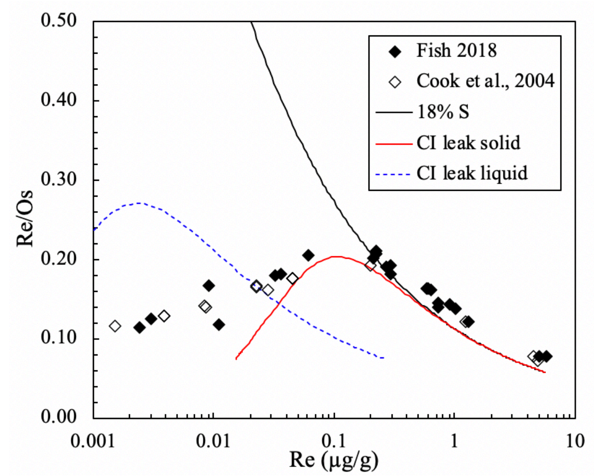

Fig. 3. Plot of Re/Os ratios against Re concentrations for IIAB iron meteorites [19, 23]. The black solid line shows Rayleigh fractionation of Re and Os for solids. The colored curves show solid (red solid line) and residual liquid (blue dashed line) tracks with a constant mixing fraction of primary melt leaked into the system.

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