

OSMIUM ISOTOPE EVIDENCE FOR AN S-PROCESS CARRIER IN PRIMITIVE CHONDRITES. A.D. Brandon¹, I.S. Puchtel², M. Humayun³, M. Zolensky⁴, ¹NASA JSC, Mail Stop KR, Houston, TX, 77058, alan.d.brandon1@jsc.nasa.gov; ²Department of Geology, University of Maryland, College Park, MD, 20742; ³National High Magnetic Field Laboratory & Department of Geological Sciences, Florida State University, 1800 E. Paul Dirac Drive, Tallahassee, Florida, 32310; ⁴NASA JSC, Mail Stop KT, Houston, TX, 77058.

Introduction: The degree of isotopic mixing in the solar nebula and the nature of pre-solar components that have contributed to our solar system remain subjects of vigorous debate. Isotopic anomalies have been identified in Ca-Al inclusions in chondrites [1-4]. This indicates that refractory pre-solar components were not completely homogenized or processed away at the high temperatures experienced by CAIs. Pre-solar grains (SiC, C, etc.) are prevalent in primitive chondrites, and preserve isotopic heterogeneity resulting from the nucleosynthetic processes occurring in the stars from which these grains formed [2,4]. Several recent studies employing precise techniques for measuring Ru, Mo and Zr isotopes in bulk meteorites, have come up with varying conclusions on the degree of effectiveness of nebular mixing on the scale of bulk meteorite material. Some of these studies have reported isotopic anomalies in Mo and Ru [3,5-7], while others have not observed anomalies in Mo, Ru, or Zr [8-10]. Debate over the quality of the data, the normalization techniques employed, the absence or presence of isobaric interferences during the measurements on different types of instruments (e.g. TIMS versus ICP-MS), and other factors, has ensued [11,12].

With these caveats in mind, we have begun a study of Os isotopes to examine issues surrounding presolar anomalies and isotopic heterogeneities in bulk meteorite samples. Osmium isotopes are produced in nucleosynthetic processes as follows: ¹⁸⁴Os is produced solely by the p-process; ¹⁸⁶Os and ¹⁸⁷Os are produced by the s-process and by radiogenic decay from ¹⁹⁰Pt and ¹⁸⁷Os, respectively; ¹⁸⁸Os, ¹⁸⁹Os, and ¹⁹⁰Os are produced dominantly by the r-process, while ¹⁹²Os is produced solely by the r-process [13-15]. Stellar nucleosynthetic processes release their products to the interstellar medium as both gas and grains. Osmium condenses as an ultrarefractory metal alloy with Re and W, and is one of the most refractory elements during condensation of stellar outflows, as long as such outflows have solar-like Os/H ratios. Ultrarefractory grains are more likely to survive nebular thermal processing, and metallic grains are less likely to be destroyed by photoelectron induced charging and confinement in magnetic loops. These factors make Os an important element to investigate for isotope anomalies. Since such effects are likely to be small, very precise measurements of Os isotopes are required. In order to search for Os isotope anomalies, we performed high-precision Os isotope measurements on unspiked aliquots of 7 ordinary chondrites (Chainpur LL3.4, Parnellee LL3.6, Sharps H3.4, St. Marguerite H3.4, Forest Vale H4, Allegan H5, Weston H5), 3 enstatite chondrites (Daniels Kuil EL6, Yilmia EL6, Indarch EH4), and 3 carbonaceous chondrites (Tagish Lake C2, Ornans CO3, Allende CV3), on the JSC Triton TIMS. Five separate fractions of the Tagish Lake chondrite were analyzed.

Analytical Technique and Results: The measured Os isotope ratios were normalized to the isotope ¹⁸⁸Os and corrected for instrumental mass fractionation using ¹⁹²Os/¹⁸⁸Os = 3.083 and the exponential law. The data are displayed in

epsilon units (Figs. 1 & 2), where ϵ_{Os} is 10^4 parts deviation from a terrestrial standard value, except for $\epsilon^{186}Os$ where the average of H-Group ordinary chondrites measured in this study was used. These ϵ_{Os} values were transformed from ¹⁸⁸Os-normalized values to ¹⁸⁹Os-normalized values adapting equation (2) of McCulloch and Wasserburg [1]. Because ¹⁸⁹Os and ¹⁹²Os are used for normalizing mass fractionation, all data have $\epsilon^{189}Os$ and $\epsilon^{192}Os$ of 0. The ¹⁸⁹Os and ¹⁹²Os isotopes were chosen for normalization because they are composed of the least amount (4.8% and 0%, respectively) of s-process component for bulk solar Os and, hence, variations in the ratio of s-process component to r-process component are most easily illustrated in this fashion. Using the traditional normalization to ¹⁸⁸Os, results in the same systematic differences between the chondrites but the patterns are shifted and s-process anomalies are obscured because of the deficiency of ¹⁸⁸Os due to s-process. To correct for radiogenic contributions from ¹⁹⁰Pt, the Pt/Os and Re/Os ratio were determined by isotope dilution ICP-MS. A fraction of each of the unspiked sample solutions was removed and separately spiked for Pt and Os concentration measurements following established techniques [16]. A multi-element spike (#000601), including ¹⁹⁰Os, ¹⁹⁸Pt, optimized for chondritic abundances, was used for the measurements. The measured Pt/Os ratios in these chondrites varied from 1.81 to 2.09, overlapping the range of a comprehensive suite of chondrites [17]. The ¹⁸⁶Os/¹⁸⁸Os ratios for the chondrites were corrected to 4.556 Ga using the Pt/Os ratios that were measured on the same solution as the high precision Os isotopic measurements were performed on. The long half-life (469 Ga) of the parent isotope, ¹⁹⁰Pt, its low abundance (0.01292 atomic%), and the restricted range in Pt/Os for chondrites, resulted in a systematic correction of all measured ¹⁸⁶Os/¹⁸⁸Os data of $1 \pm 0.1 \epsilon^{186}Os$ units. The ¹⁸⁷Os ratios are dominated by radiogenic ingrowth of ¹⁸⁷Re, and are not shown on the figures. The p-process isotope, ¹⁸⁴Os, is too low in abundance (0.02%) to measure with sufficient precision and is not plotted.

When normalized to the r-process dominated isotopes, ¹⁸⁹Os and ¹⁹²Os, Tagish Lake (C2), and to a lesser extent Ornans (CO3), display deficiencies in the s-process isotopes relative to the majority of the ordinary and enstatite chondrites measured with anomalies in the order: ¹⁸⁶Os > ¹⁸⁸Os > ¹⁹⁰Os (Figs. 1 & 2). Interestingly, these deficiencies qualitatively correlate with the s-process abundances of each isotope (Fig. 2). Also, Parnellee (LL3.6) and Indarch (EH4) exhibit a small negative $\epsilon^{186}Os$ (100% s-process isotope) anomaly.

Discussion and Conclusions: The variations in ϵ_{Os} observed could result from 3 processes: cosmic-ray exposure effects, incomplete digestion of insoluble Os carrying phases, or inhomogeneous distribution of the s/r process isotopes in the solar nebula. Each of these processes would have important implications on isotopic heterogeneity in chondrites in relation to nebular heterogeneity.

The Os isotopic compositions of H-Group ordinary chondrites are indistinguishable from the terrestrial standard and terrestrial upper mantle samples. Hence, the only

mechanism that can cause this relationship from cosmic-ray exposure is consumption of ^{186}Os and to a lesser extent, ^{188}Os and ^{190}Os during cosmic-ray bombardment in the samples with isotopic depletions relative to the H-Group chondrites. Leya (personal communication 2004) has estimated the effects on Os isotopes from cosmic-ray exposure (following [18]). For an exposure age of 100 Ma, the effect should be from 0.2 to 1 epsilon units for burnout of the different Os isotopes. The exposure age for Tagish Lake calculated from ^{21}Ne is 7.8 Ma [19], and hence, the effects for cosmic ray exposure should be much less than what is shown in Fig. 2.

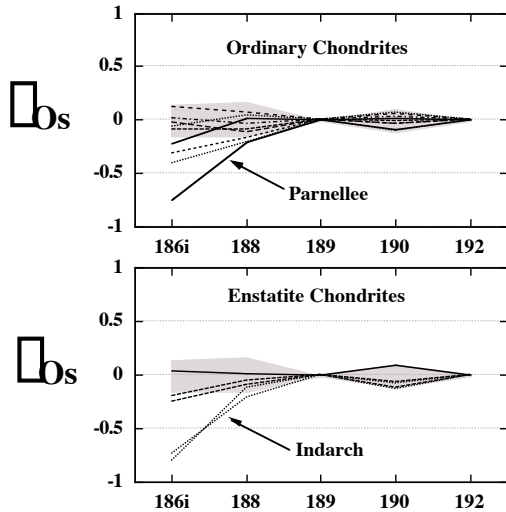


Figure 1. Os isotopic compositions of H-Group (upper diagram dashed lines), LL-Group (upper diagram solid lines), and E-chondrites (lower diagram). The shaded region in these diagrams is the $\pm 2\sigma$ error envelope on 39 runs of the terrestrial standard. Terrestrial mantle samples [20] all plot within this envelope. Individual data points for each sample have $\pm 0.2 \epsilon_{\text{Os}}$ (2σ) or better uncertainties.

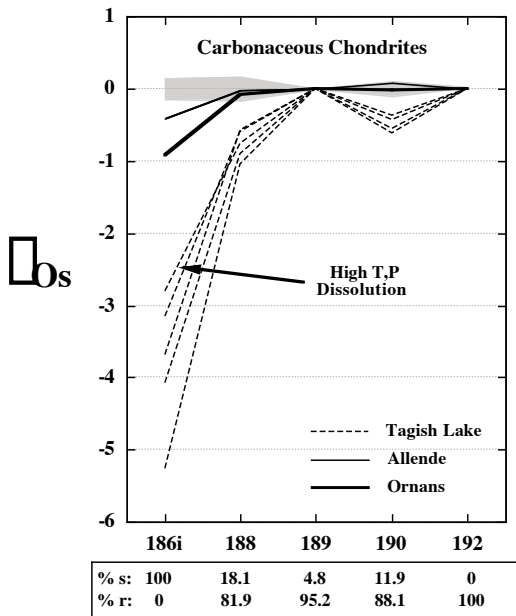


Figure 2. Carbonaceous chondrite Os isotopic data.

Osmium is hosted in Fe-Ni alloy, refractory metal nuggets, and possibly in presolar SiC or graphite, similar to Ru. To determine whether incomplete digestion of an Os-carrier was responsible for the effect, a digestion of Tagish Lake was performed using sealed metal jackets with CO_2 overpressure, at temperatures of 325°C (following [21]), relative to $240\text{-}270^\circ\text{C}$ for the other digestions. The Os isotopic data for this digestion retains the Os isotope anomalies (Fig. 2). In addition, the five separate digestions of different clasts from the Tagish Lake meteorite show variable anomalies with systematic depletions in $^{186}\text{Os} > ^{188}\text{Os} > ^{190}\text{Os}$. These observations imply an s-process deficit in Tagish Lake, and suggest that whatever phase(s) in Tagish Lake is carrying the s-process Os, it is either heterogeneously distributed or variably accessed by the digestions. From the s-process anomalies observed in Tagish Lake, the estimated amount of s-process Os deficit (20 pg/g) is about 40×10^{-6} of the total Os (476 ng/g). If the carrier phase, perhaps SiC, constitutes about 1-10 ppm by weight of Tagish Lake, then the s-process Os concentration is about 2-20 ppm. Actual Os concentrations could be higher due to isotopically normal Os being incorporated from the stellar atmosphere. Elemental abundances in presolar SiC by SXRF for Ru (2-57 ppm) and Mo (1-54 ppm) [22] are of the same order of magnitude as the mean Os abundance (2-20 ppm) in the s-process carrier inferred here. The chemical similarity of Os to Ru indicates that some Os may occur in SiC, even though neither element is known to form carbides. Osmium may occur as refractory metal inclusions within other presolar grains.

Hence, the most likely interpretation of these data is that an undigested carrier phase retains the s-process Os required to yield the solar s/r ratio for Os. Such a phase is not expected to be preserved in higher metamorphic grade chondrites. Thus, further studies of metamorphosed C chondrites, and primitive O and E chondrites should test this hypothesis. We will concentrate, and then attempt to extract, the s-process Os carrier(s) in future work.

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