



## s-Process Os isotope enrichment in ureilites by planetary processing



S. Goderis<sup>a,b,c,\*</sup>, A.D. Brandon<sup>c</sup>, B. Mayer<sup>d</sup>, M. Humayun<sup>d</sup>

<sup>a</sup> Earth System Science, Vrije Universiteit Brussel, Brussels, Belgium

<sup>b</sup> Department of Analytical Chemistry, Ghent University, Ghent, Belgium

<sup>c</sup> Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, USA

<sup>d</sup> National High Magnetic Field Laboratory and Department of Earth, Ocean & Atmospheric Science, Florida State University, Tallahassee, FL, USA

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### ABSTRACT

Ubiquitous nucleosynthetic isotope anomalies relative to the terrestrial isotopic composition in Mo, Ru, and other elements are known from both bulk chondrites and differentiated meteorites, but Os isotope ratios reported from such meteorites have been found to be indistinguishable from the terrestrial value. The carriers of s- and r-process Os must thus have been homogeneously distributed in the solar nebula. As large Os isotope anomalies are known from acid leachates and residues of primitive chondrites, the constant relative proportions of presolar s- and r-process carriers in such chondrites must have been maintained during nebular processes. It has long been assumed that partial melting of primitive chondrites would homogenize the isotopic heterogeneity carried by presolar grains. Here, ureilites, carbon-rich ultramafic achondrites dominantly composed of olivine and low-Ca pyroxene, are shown to be the first differentiated bulk Solar System materials for which nucleosynthetic Os isotope anomalies have been identified. These anomalies consist of enrichment in s-process Os heterogeneously distributed in different ureilites. Given the observed homogeneity of Os isotopes in all types of primitive chondrites, this Os isotope variability among ureilites must have been caused by selective removal of s-process-poor Os host phases, probably metal, during rapid localized melting on the ureilite parent body. While Mo and Ru isotope anomalies for all meteorites measured so far exhibit s-process deficits relative to the Earth, the opposite holds for the Os isotope anomalies in ureilites reported here. This might indicate that the Earth preferentially accreted olivine-rich restites and inherited a s-process excess relative to smaller meteorite bodies, consistent with Earth's high Mg/Si ratio and enrichment of s-process nuclides in Mo, Ru, and Nd isotopes. Our new Os isotope results imply that caution must be used when applying nucleosynthetic isotope anomalies as provenance indicators between different classes of meteorites.

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### 1. Introduction

Isotopic anomalies are defined as deviations from terrestrial or bulk solar isotopic abundances that have resulted from processes other than radioactive decay or mass-dependent fractionation. Presolar grains, widespread in unequilibrated chondrites, are enriched in material derived from distinct nucleosynthetic environments (Zinner, 1998). Small but resolvable isotopic anomalies in bulk meteorites, often most prominent in primitive carbonaceous chondrites, are present for various elements including Ti, Cr, Ni, Zr, Mo, Ru, Ba, Nd, and Sm (e.g., Akram et al., 2015; Burkhardt et al., 2011, 2012; Carlson et al., 2007; Chen et al., 2010;

Dauphas et al., 2002; Qin et al., 2010; Regelous et al., 2008; Trinquier et al., 2009; Yin et al., 2002). Uniform isotopic compositions in bulk chondrites are reported for other elements, most prominently Os and Hf (Brandon et al., 2005; Sprung et al., 2010; van Acken et al., 2011; Walker, 2012; Wittig et al., 2013; Yokoyama et al., 2007). The presence of isotope anomalies in bulk planetary materials, such as chondrites and achondrites, is explained by a variety of mechanisms that resulted in isotope heterogeneity at the planetesimal scale. These include, for example, inheritance from a heterogeneous molecular cloud core, formation of localized heterogeneities due to differential physical sorting of dust grains (e.g., metals, silicates and sulfides), late supernova injection, or selective destruction of thermally labile presolar carrier phases in the nebula (e.g., Andreasen and Sharma, 2007; Burkhardt et al., 2011; Carlson et al., 2007; Chen et al., 2010; Dauphas et al., 2002; Qin et al., 2008; Trinquier et al., 2009). Alternatively, secondary processes, such as aqueous alteration have been suggested to selec-

\* Corresponding author at: Earth System Science, Analytical, Environmental, and Geo-Chemistry (AMGC), Vrije Universiteit Brussel, Pleinlaan 2, B-1050 Brussels, Belgium. Tel.: +32 26291480.

E-mail address: Steven.Goderis@vub.ac.be (S. Goderis).

tively redistribute isotopically distinct components within a planetesimal (Yokoyama et al., 2011).

Osmium is an important element in this discussion, as it is one of the most refractory metals. Chemical residues and leachates of various chondrites of low metamorphic grade show complementary Os isotope anomalies, attributed to the presence of varying proportions of presolar grains carrying s-, r-, and possibly p-process Os (Brandon et al., 2005; Reisberg et al., 2009; van Acken et al., 2011; Yokoyama et al., 2007, 2010, 2011). Analyses of bulk chondrites using total digestion methods, thermally metamorphosed chondrites, and iron meteorites, however, have shown no evidence for resolvable Os isotopic heterogeneity within the current level of analytical resolution (Brandon et al., 2005; van Acken et al., 2011; Walker, 2012; Wittig et al., 2013; Yokoyama et al., 2007, 2010, 2011). This indicates that, although carriers with distinct Os isotopic compositions were present in the protosolar nebula, their relative proportions were fixed in all primitive chondrites, consistent with a well-mixed solar nebula (Brandon et al., 2005; Sprung et al., 2010; Yokoyama et al., 2007; Walker, 2012). Since low-grade metamorphism is sufficient to destroy presolar grains (Huss and Lewis, 1995), it has been assumed that partial melting should eliminate any small-scale Os isotope anomalies in the sources of differentiated bodies. The existing high-precision Os isotope data indicate that iron meteorites have the same Os isotopic composition as chondrites, consistent with homogenization of isotopically diverse components during melting and core formation (Walker, 2012; Wittig et al., 2013).

Ureilites are coarse-grained ultramafic achondrites, consisting primarily of olivine and pigeonite, with minor augite and orthopyroxene, abundant carbon (<7 wt%, graphite and diamond) and accessory metal and sulphides (e.g., Barrat et al., 2015; Goodrich et al., 2004; Warren et al., 2006). Ureilites are widely considered to represent mantle restites, formed after the extraction of silicate partial melts and S-rich metallic melts (e.g., Goodrich et al., 2004; Warren and Kallemeyn, 1992). Highly siderophile elements confirm that metallic liquids were extracted from ureilites with most ureilite metal having enrichments in compatible siderophile elements (Goodrich et al., 2013; Horstmann et al., 2014; Rankenburg et al., 2008). The heavy Fe isotopic composition of ureilites compared to chondrites corroborates an efficient segregation of S-rich metallic melts in the ureilite body, likely before the onset of silicate melting, and is interpreted to represent the early stages of asteroidal core formation (Barrat et al., 2015). Despite evidence of an igneous origin, ureilites also appear to have preserved primordial signatures in light stable isotopes. The ureilites do not follow a unique mass-dependent isotopic fractionation trend in oxygen isotope space (Clayton and Mayeda, 1996). Instead, ureilites trend along the carbonaceous chondrite anhydrous minerals (CCAM) line (Supplementary Fig. 1). Ureilites exhibit  $^{33}\text{S}$  isotope enrichment (Farquhar et al., 2000), and large differences in  $\delta^{15}\text{N}$  between diamond, graphite, and silicate phases (Rai et al., 2003a). These isotopic heterogeneities likely reflect primordial nebular heterogeneity of the accreting precursor materials and thus argue against magmatic equilibration of the ureilite parent body (UPB). Isotopic data for Cr, Ti, and Ni for ureilites (e.g., Regelous et al., 2008; Trinquier et al., 2009) add to the enigma of ureilites, as the reported  $\varepsilon^{50}\text{Ti}$ ,  $\varepsilon^{54}\text{Cr}$ , and  $\varepsilon^{62}\text{Ni}$  (with the  $\varepsilon$ -notation representing isotopic deviations from a reference value in parts per 10,000) show the lowest (negative) values of all meteorites, grouping far from the carbonaceous chondrites that are themselves separated by a wide margin from all other planetary materials (Warren, 2011).

Following the earlier work of Rankenburg et al. (2007, 2008) on highly siderophile element abundances and  $^{187}\text{Os}/^{188}\text{Os}$  isotope ratios, we obtained the first high-precision Os isotope data for a suite of mostly monomict ureilites using negative thermal ionization

mass spectrometry (N-TIMS). This is the first high-precision isotope data obtained for ureilites for elements beyond the Fe-peak. Isotope dilution concentration data for highly siderophile elements (HSE: Ru, Pd, Re, Os, Ir, and Pt) are also reported on the same samples, to correct for radiogenic contributions from  $^{187}\text{Re}$  and  $^{190}\text{Pt}$  to  $^{187}\text{Os}$  and  $^{186}\text{Os}$ , respectively. Serendipitously, the high-precision Os isotope data revealed nucleosynthetic isotope anomalies. These data are used to place constraints on the genesis and early evolution of UPB reservoirs. Studying the Os isotopic signatures of ureilites provides a way to test possible large-scale heterogeneity linked to the inhomogeneous distribution of presolar material in the nascent solar nebula, and to evaluate the effects of planetary processes, such as partial melting, on the distribution of the carriers of nucleosynthetic isotope anomalies during planetary differentiation.

## 2. Materials and methods

Twelve monomict ureilite breccia samples were obtained from the Smithsonian Institution's National Museum of Natural History (Goalpara, Kenna), the NASA Meteorite Work Group (ALHA 81101, EET 87517, GRA 95205, GRA 98032, META 78008), and the Japanese National Institute of Polar Research (A 881931, Y 790981, Y 791538, Y 981750, Y 981810). Libyan Desert find DaG 319, the only polymict ureilite studied in this work, was purchased from a commercial meteorite dealer. Prior to dissolution, all surfaces on all pieces of meteorites were polished using carborundum, after which the meteorites were manually ground with a ceramic alumina mortar and pestle. In the methodology applied, high-precision determination of Os isotope ratios on unspiked digestions (Supplementary Table 1; Table 1) is combined with determination of elemental abundance ratios of Re, Os, Ir, Ru, Pt, and Pd, on small aliquots taken from the unspiked digestions to ensure the representativeness of these ratios for each sample digestion. In this way,  $^{186}\text{Os}$  and  $^{187}\text{Os}$  contributions generated by the long-lived radioactive  $^{187}\text{Re}$ – $^{187}\text{Os}$  and  $^{190}\text{Pt}$ – $^{186}\text{Os}$  decay systems with half-lives of 41.5 Ga and 488 Ga, respectively (Begemann et al., 2001; Smoliar et al., 1996), can be taken into account. Elemental ratios of interest are also checked by comparison to previously published data on the same meteorites (Table 2 and Fig. 1; Rankenburg et al., 2007, 2008). In order to obtain the amount of Os required for the high-precision Os isotope ratio measurements (~100 ng), each sample was digested in one to six curius tubes (CTs), depending on the Os concentration in the sample (ALHA 81101 and A 881931 with <80 ppb Os were digested in 6 and 3 CTs, respectively). For the initial unspiked digestions, ~0.5 g of sample powder was digested in acid leached borosilicate glass CTs for 48 h at 230 °C with 3 ml conc. HCl and 6 ml conc. HNO<sub>3</sub>. After that, the tubes were frozen and opened, and 0.45 ml of the acid sample solution transferred into a quartz CT for precise determination of elemental abundances of the ureilites. After an aliquot of digested sample solution was transferred, the quartz CT was chilled and an appropriate amount of mixed  $^{99}\text{Ru}$ – $^{110}\text{Pd}$ – $^{185}\text{Re}$ – $^{190}\text{Os}$ – $^{191}\text{Ir}$ – $^{198}\text{Pt}$  spike (#000601) was added, followed by 1 ml of conc. HCl and 2 ml of conc. HNO<sub>3</sub>. Sample-spike equilibration was achieved in an oven at 230 °C for 24 h after sealing of the CTs. Where samples were split across several CTs, the Pt/Os ratios obtained for one of these CTs were compared either to literature data (ALHA 81101) or duplicate analysis of the same sample powder (A 881931) and found indistinguishable within uncertainty (Table 2). Regardless,  $^{190}\text{Pt}/^{188}\text{Os}$  radiogenic corrections were found to be marginal compared to the observed nucleosynthetic effects (Table 1, Supplementary Table 1).

Unspiked aliquot Os from the batch of CTs containing a single sample digestion was extracted using CCl<sub>4</sub>/HBr solvent extraction (Cohen and Waters, 1996), subsequently purified using

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