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Intrinsic W nucleosynthetic isotope variations in carbonaceous chondrites: Implications for W nucleosynthesis and nebular vs. parent body processing of presolar materials

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Abstract

The progressive dissolution of the carbonaceous chondrites Orgueil (CI1), Murchison (CM2) and Allende (CV3) with acids of increasing strength reveals correlated W isotope variations ranging from 3.5 $\varepsilon^{182}W$ and 6.5 $\varepsilon^{183}W$ in the initial leachate (acetic acid) to −60 $\varepsilon^{182}W$ and −40 $\varepsilon^{183}W$ in the leachate residue. The observed variations are readily explained by variable mixing of *s*-process depleted and *s*-process enriched components. One W *s*-process carrier is SiC, however, the observed anomaly patterns and mass–balance considerations require at least on additional *s*-process carrier, possibly a silicate or sulfide. The data reveal well-defined correlations, which provide a test for *s*-process nucleosynthesis models. The correlations demonstrate that current models need to be revised and highlight the need for more precise W isotope data of SiC grains. Furthermore the correlations provide a mean to disentangle nucleosynthetic and radiogenic contributions to ^{182}W ($\varepsilon^{182}W_{corrected} = \varepsilon^{182}W_{measured} - (1.41 \pm 0.05) \times \varepsilon^{183}W_{measured}$; $\varepsilon^{182}W_{corrected} = \varepsilon^{182}W_{measured} - (-0.12 \pm 0.06) \times \varepsilon^{184}W_{measured}$), a prerequisite for the successful application of the Hf–W chronometer to samples with nucleosynthetic anomalies.

The overall magnitude of the W isotope variations decreases in the order CI1 > CM2 > CV3. This can be interpreted as the progressive thermal destruction of an initially homogeneous mixture of presolar grains by parent-body processing. However, not only the magnitude but also the W anomaly patterns of the three chondrites are different. In particular leach step 2, that employs nitric acid, reveals a *s*-deficit signature for Murchison, but a *s*-excess for Orgueil and Allende. This could be the result of redistribution of anomalous W into a new phase by parent-body alteration, or, the fingerprint of dust processing in the solar nebula. Given that the thermal and aqueous alteration of Murchison is between the CI and CV3 chondrites, parent-body processing is probably not the sole cause for creating the different pattern. Small-scale nebular redistribution of anomalous W may have played a role as well. Similar nebular processes possibly acted differently on specific carrier phases and elements, resulting in the diverse nucleosynthetic signatures observed in planetary materials today.

1. INTRODUCTION

Chondritic meteorites are space sediments whose various components were produced in stellar, nebular and parent body environments (Krot et al., 2009). Isotope anomalies of nucleosynthetic origin identified for bulk rock chondrites and their components can provide key constraints on many of the involved processes and

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environments, such as stellar nucleosynthesis, physicochemical processing and mixing of matter in the solar circumstellar disk, planetary body accretion and parent body metamorphism.

Nucleosynthetic variations in meteorites are observed on three different scales. The largest variations (isotopic deviations relative to terrestrial or average solar system composition in the % range or higher) are obtained by the direct measurements of single presolar grains isolated from the matrix of non-equilibrated chondritic meteorites (Zinner, 2014). These grains condensed around dying stars and provide a direct probe to nucleosynthetic processes occurring in their host stars and thus are invaluable samples to test and refine theoretical models of nucleosynthesis. However, these types of measurements are intricate, often associated with large uncertainties, limited to specific types of presolar grains (mainly SiC) and until now only available for certain elements (*e.g.*, Ott and Begemann, 1990; Nicolussi et al., 1997,1998; Lugaro et al., 2003; Podosek et al., 2004; Avila et al., 2012). The presence of presolar grains in chondritic meteorites provides unequivocal evidence that some primitive materials from the solar systems parental cloud survived nebular processing (Lewis et al., 1987). This implies that the solar nebula was never in complete isotopic equilibrium and thus a pure condensation origin can be excluded for most chondrite components.

Presolar grains have also been used as a proxy for parent body alteration within and between chondrite classes (*e.g.*, Huss and Lewis, 1995; Davidson et al., 2014). The findings show that the lower the amount of presolar grains within a specific chondrite of the same chondrite class, the higher is the degree of parent body alteration experienced by the meteorite. Comparison of presolar grain abundances between chondrite classes, however, is less straightforward. This is because the various presolar phases can react differently to alteration if fundamental conditions change, *e.g.*, at a given temperature SiC seems to be more stable in the reducing conditions of enstatite chondrites than in the oxidizing environment of carbonaceous chondrites (Huss and Lewis, 1995). Furthermore, as the various chondrite classes formed under different nebular (redox) conditions and vary in composition and texture, they might also sample variably processed blends of presolar materials. Therefore, the variable presolar grain abundances in the different chondrite classes might not only be due to different degrees of parent body alteration, but also indicative of varying pre-accretionary material processing in the solar nebula (Huss et al., 2003). Such processing could lead to a heterogeneous distribution of presolar materials in the solar system and might be responsible for establishing nucleosynthetic isotope variations on a planetary scale.

Planetary-scale nucleosynthetic isotope variations, that are reported for bulk chondrites and differentiated meteorites, are small (isotopic variations relative to terrestrial compositions are in the parts per 10^4 to parts per 10^6 range), but well resolved and established for a growing number of elements (*e.g.*, Ca, Ti, Ni, Cr, Sr, Zr, Mo, Ru, Nd, Sm) (Dauphas et al., 2002b; Andreasen and Sharma, 2007; Trinquier et al., 2007, 2009; Regelous et al., 2008; Burkhardt et al., 2011; Chen et al., 2010, 2011; Gannoun et al., 2011; Moynier et al., 2012; Akram et al., 2013, 2015). These bulk-scale anomalies are used to infer genetic relations between meteorite parent bodies and bear witness to a large-scale nebular isotopic heterogeneity that can help to better constrain material processing and mixing dynamics in the early solar system. Several models have been put forward to explain the isotopic heterogeneity among bulk meteorites. The heterogeneity could (i) reflect a primordial feature of the solar nebula inherited from a large scale heterogeneous parental molecular cloud (Clayton, 1982; Dauphas et al., 2002b), (ii) be caused by the injection of isotopically heterogeneous matter into the nebula (Lee et al., 1977), or (iii) is the result of physical and/or chemical dust processing within an initially homogeneous nebula such as *e.g.*, grain size sorting (Dauphas et al., 2010), grain type sorting (Regelous et al., 2008) or selective destruction of thermally labile presolar components in different nebular environments (Trinquier et al., 2009; Burkhardt et al., 2012a). Although the latter model is currently favored because it allows for the generation of bulk scale isotopic anomalies for some elements *and* uniform isotopic composition for others, the specific nebular processes involved are not well understood yet.

The progressive dissolution of chondritic meteorites with acids of increasing strength reveals the internal nucleosynthetic variability of chondrites. These experiments yield precise isotopic data that scale between those of single presolar grain data and those of the bulk meteorites (isotopic variations relative to terrestrial compositions are in the ‰ to parts per 10^4 range). Currently, leachate data are available for a number of primitive chondrites (mainly carbonaceous, but also some enstatite and ordinary chondrites) and for a variety of elements including Ti, Cr, Sr, Zr, Mo, Te, Ba, Nd, Sm, W and Os (*e.g.*, Rotaru et al., 1992; Dauphas et al., 2002a; Hidaka et al., 2003; Schönbächler et al., 2003, 2005; Fehr et al., 2006; Trinquier et al., 2007, 2009; Reisberg et al., 2009; Yokoyama et al., 2010, 2011; Qin et al., 2011; Burkhardt et al., 2012a,b; Boyet and Gannoun, 2013). The leaching technique represents a quick way to establish precise nucleosynthetic isotope data for a range of elements and provide constraints on the carrier phases of the nucleosynthetic variations. The information obtained from leachates is used to test and refine nucleosynthesis models, unravel the processes that lead to the formation of bulk-scale anomalies in some elements but not in others, and address the question of nebular vs. parent body alteration. Furthermore, leachate data are needed to adequately correct nucleosynthetic anomalies in planetary materials, a prerequisite for the successful applications of some short-lived chronometers (Qin et al., 2011; Burkhardt et al., 2012b).

Previous W leachate data is limited to the carbonaceous chondrite Murchison (Burkhardt et al., 2012b). In this study, we extended this data and present W isotope data for acid leachates of the carbonaceous chondrites Orgueil (CI1), Murchison (CM2) and Allende (CV3). Aliquots from the same leach fractions were previously analyzed for Zr (Schönbächler et al., 2005) and Te (Fehr et al., 2006). The new data provide insight into nature and origin of the nucleosynthetic W isotope variations in our solar system