

NUCLEOSYNTHETIC Mo ISOTOPE ANOMALIES IN ACID LEACHATES OF THE MURCHISON CHONDRITE AND THEIR RELEVANCE FOR EARLY SOLAR SYSTEM PROCESSES. C. Burkhardt¹, T. Kleine², N. Dauphas³, F. Oberli¹ and R. Wieler¹. ¹Institute of Geochemistry and Petrology, Clausiusstrasse 25, ETH Zürich NW D84, CH-8092 Zürich (burkhardt@erdw.ethz.ch). ²Institut für Planetologie, Westfälische Wilhelms-Universität Münster, Wilhelm-Klemm Strasse 10, D-48149 Münster. ³Origins Laboratory, Dept. of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, 5734 South Ellis Avenue, Chicago, IL 60637.

Introduction: The discovery of presolar grains in primitive meteorites some 30 years ago demonstrated that the longstanding assumption of a hot, isotopically homogeneous solar nebula [1] is invalid and that at least a fraction of the nebula's dust escaped high-temperature processing within the solar nebula. A question of great importance for understanding the early evolution of the solar nebula is as to whether this presolar, isotopically anomalous dust has been well mixed in the solar nebula prior to the onset of planetary accretion. In recent years a growing number of studies reported nucleosynthetic anomalies in bulk meteorites [e.g. 2-5], indicating that different planetary bodies incorporated varying proportions of presolar dust.

However, the planetary-scale heterogeneity documented for some elements (e.g. Cr, Ti, Mo) contrasts with the absence of any evidence for such heterogeneities for other elements (e.g. Os). Understanding such disparate behavior requires knowledge regarding the carrier phases that host the isotopic anomalies. Such information can be obtained by the stepwise dissolution of chondrites. We here present Mo isotopic data for acid leachates of the Murchison chondrite that were previously investigated for their Os isotopic compositions. The combined Mo-Os data set allows assessing the causes for the observed planetary scale isotopic heterogeneity for Mo and its absence for Os.

Methods: Aliquots from leachates and the insoluble residue prepared from ~16.5 g powdered Murchison at the University of Chicago were analyzed. The leaching sequence was as follows (for details see [6]):

- L1: 9M HAc, 1 day, 20 °C;
- L2: 4.7M HNO₃, 5 days, 20 °C;
- L3: 5.5M HCl, 1 day, 75 °C;
- L4: 13M HF 3M HCl, 1 day, 75 °C;
- L5: 13M HF 6M HCl, 3 days, 150 °C;
- L6: insoluble residue.

After drying down the leachates were treated with aqua regia, dried and then redissolved in HCl. The insoluble residue remaining after five leaching steps (L6) was fused by a CO₂ laser under a reducing atmosphere and then digested in HNO₃-HF-HClO₄. Mo concentrations in the leachates and the residue were determined by isotope dilution on small aliquots. Purification of Mo from the sample matrix followed our previously established techniques [3]. Mo isotopic composi-

tions were determined using the Nu Plasma 1700 MC-ICP-MS at ETH Zürich and are reported in $\epsilon^i\text{Mo}$ as the part per 10,000 deviation from the terrestrial Mo isotopic composition. Instrumental mass bias was corrected relative to $^{98}\text{Mo}/^{96}\text{Mo}=1.453174$ using the exponential law. Isobaric Zr and Ru interferences were monitored using ^{90}Zr and ^{99}Ru and were negligible.

Results: Mo isotope data for leachates L1-L5 and the acid residue (L6) are shown in Fig. 1 and 2.

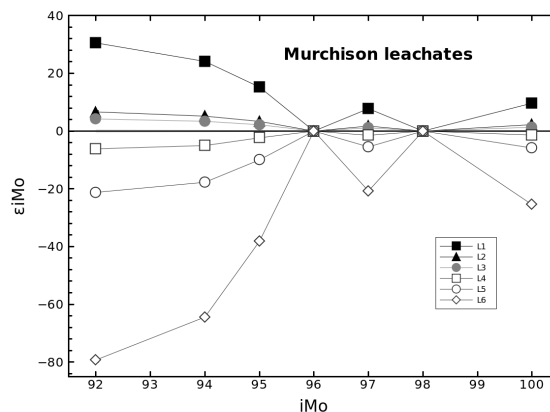


Figure 1: Mo isotopic compositions of Murchison leachates (L1-L5) and acid-resistant residue (L6) in a ^4Mo vs. $\epsilon^1\text{Mo}$ plot.

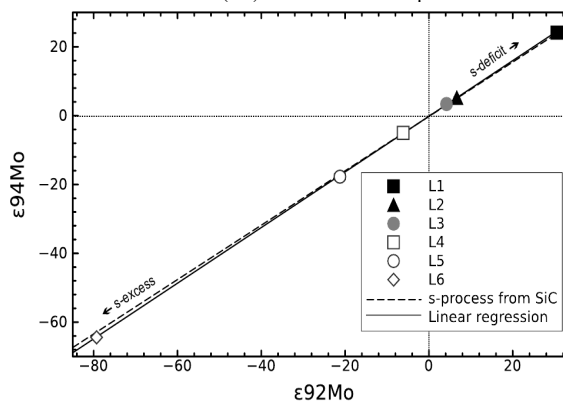


Figure 2: Plot of $\epsilon^{92}\text{Mo}$ vs. $\epsilon^{94}\text{Mo}$. All samples plot on a mixing line between pure s-process Mo [7] and terrestrial Mo ($\epsilon^i\text{Mo}=0$).

All the acid leachates and the residue have Mo isotopic compositions different from the terrestrial value (Fig. 1). The observed isotope patterns can be accounted for by variable amounts of s-process derived Mo in the investigated Murchison samples (Fig. 2). While L1- L3 exhibit a deficit in s-process Mo isotopes relative to the Earth, L4-L6 show an s-excess. The com-

bined data for L1 to L6 yield a Mo concentration of $\sim 1.1 \mu\text{g/g}$ and a Mo isotopic composition corresponding to an s-process deficit relative to the Earth ($\sim -4.5 \epsilon^{92}\text{Mo}$). Both the Mo concentration and isotopic compositions are consistent with an independent determination on a bulk sample of Murchison [3].

Discussion: Mo isotope anomalies in acid leachates were previously investigated for Orgueil and Allende [8], using a slightly different leaching protocol compared to the one used here. For Orgueil the pattern of Mo isotope anomalies is broadly similar to that observed for Murchison with two important differences: (i) in Murchison the largest s-process deficit is released in HAc (L1), but no Mo isotope anomaly was found in the HAc leachate of Orgueil. Either this easily leachable and highly anomalous component was not present in Orgueil or it has been destroyed during alteration on the parent body; (ii) the residue of Murchison (L6) exhibits a large s-process excess ($\sim -80 \epsilon^{92}\text{Mo}$) but the residue of Orgueil shows no Mo isotope anomaly, perhaps because it was not completely digested. Alternatively this s-process enriched component was not a significant fraction of the parental material of CI chondrites. For Allende all leaching steps show identical and slightly positive anomalies that are indistinguishable from the bulk. The comparison of the Mo isotope results for acid leachates of the three different carbonaceous chondrites reveals that Murchison (i) contains the components with the largest Mo isotope anomalies yet found, and (ii) preserved the largest spread in Mo isotopic compositions. As to whether this reflects a primary feature of the material accreted to the CM chondrite parent body or is the result of dilution of the anomalies by parent body processes on the CI and CV parent bodies is unclear. In any case, Murchison appears to be ideally suited to study the primordial variability of Mo isotopes in planetary building blocks.

Nucleosynthetic anomalies in acid leachates of Murchison were investigated for several other elements, including Zr [9] and Os [6]. Variations in ^{96}Zr abundances in Murchison leachates reflect depletions and enrichments in s-process material consistent with those observed here for Mo (Fig. 3): the early leaching steps (L1-3) are characterized by a depletion in s-process isotopes while the later steps (L4-6) reveal s-process enrichment. As Zr and Mo have different chemical behaviors, the coherent Zr and Mo isotopic signatures in the leachates indicates that Zr and Mo are hosted in the same carrier(s). This is not surprising as these two neighboring elements were probably formed in the same nucleosynthetic event(s). The correlated appearance of Mo and Zr isotopic anomalies in the Murchison leachates combined with the observation of

widespread Mo isotopic anomalies in bulk meteorites [2,3] implies that bulk meteorites should have Zr isotopic anomalies. However, no such anomalies have yet been found [9], perhaps because the precision of the Zr isotope data achievable at the time was insufficient to resolve the small anomalies in the bulk meteorites.

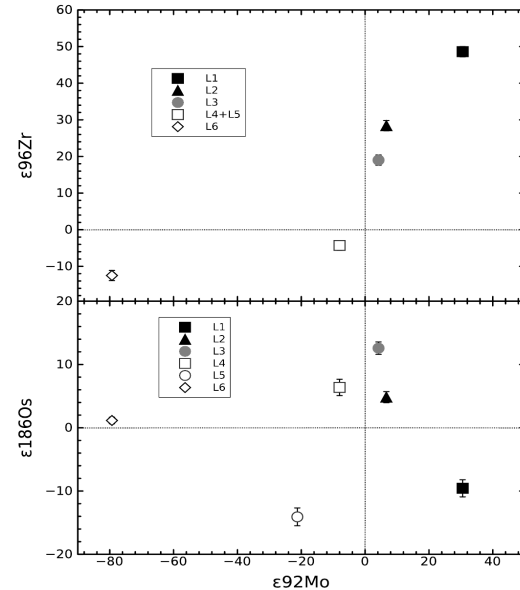


Figure 3: Zirconium [9], Os [6] and Mo (this study) isotopic compositions of Murchison leachates (L1-L5) and insoluble residue (L6).

Osmium isotopic data were determined in a previous study [6] for aliquots from the very same leachates used in this study, permitting a direct comparison of nucleosynthetic Mo and Os isotope anomalies. Fig. 3 reveals that the Mo and Os isotopic anomalies do not correlate: L1 and L5 show a deficit in s-process Os isotopes (or an excess in r-process isotopes), L2-L4 exhibit a s-excess (r-deficit), and L6 shows only a very small s-excess (r-deficit). For all samples except L1 the nucleosynthetic signature is different for Mo and Os, indicating that these two elements must reside in different carrier phases. This can account for the observation that nucleosynthetic isotopic anomalies exist in bulk meteorites for Mo [2,3] but not for Os [10]. Obviously the carrier(s) of the different Os isotopes were efficiently mixed in the solar nebula, while those of Mo isotopes were heterogeneously distributed.

References: [1] Suess H. E. (1965) *ARA&A*, 3, 217-234. [2] Dauphas N. et al. (2002) *ApJ*, 565, 640-644. [3] Burkhardt C. et al. (2011) *this volume*. [4] Trinquier A. (2009) *Science*, 324, 374-376. [5] Chen J. et al. (2010) *GCA*, 74, 3851-3862. [6] Reisberg L. et al. (2009) *EPSL*, 277, 334-344. [7] Lugaro M. et al. (2003) *ApJ*, 593, 486-508. [8] Dauphas N. et al. (2002) *ApJ*, 569, 139-142. [9] Schönbacher M. et al. (2005) *GCA*, 69, 5113-5122. [10] Yokoyama T. et al. (2007) *EPSL*, 259, 567-580.