

**IRON AND SELENIUM ISOTOPE HOMOGENEITY IN THE PROTOSOLAR NEBULA?** N. Dauphas<sup>1,2</sup>, O. Rouxel<sup>3</sup>, A. M. Davis<sup>1</sup>, R.S. Lewis<sup>1</sup>, M. Wadhwa<sup>2</sup>, B. Marty<sup>4</sup>, L. Reisberg<sup>4</sup>, P. E. Janney<sup>2</sup>, and C. Zimmermann<sup>4</sup>, <sup>1</sup>Enrico Fermi Institute, The University of Chicago, 5640 South Ellis Avenue, Chicago, IL 60637, USA (dauphas@uchicago.edu), <sup>2</sup>Department of Geology, The Field Museum, 1400 South Lake Shore Drive, Chicago, IL 60605, USA, <sup>3</sup>Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge CB2 3EQ, UK, <sup>4</sup>CRPG, 15 rue Notre Dame des Pauvres, BP 20, 54501 Vandoeuvre lès Nancy Cedex, France.

**Introduction:** At a microscopic scale, the isotopic compositions of most elements are heterogeneous. Indeed, meteorites host circumstellar grains that survived the planetary scale isotope homogenization associated with collapse of the protosolar nebula [1]. The composition of these grains is that of the stellar environment where they formed and the isotopic variations often must be displayed with logarithmic scales. At the other extreme, at a planetary scale, a few elements still display some heterogeneity limited to at most a few permil [2-7]. As the precision in isotope determinations increases, an increasing number of elements provide evidence for such heterogeneity. Identification of isotope anomalies may help identify new presolar phases, allow better understanding of nebular processes such as gas-dust decoupling and granular sorting, and help to trace genetic relationships between solar system objects. Leaching experiments in primitive meteorites have proven to be very useful for revealing nucleosynthetic anomalies [7-10]. The isotopic composition of molybdenum was measured recently in leachates of carbonaceous chondrites [7]. A dichotomy was observed between *s* and *pr* nuclides. This dichotomy probably results from the presence of a presolar phase bearing an *s*-process signature because *p* and *r* anomalies are not decoupled. In the present contribution, we pursue this work and report the determination of selenium and iron isotope abundances in these leaching experiments.

**Iron isotopes:** Isotope anomalies in leaching experiments were first identified for chromium [8, 9]. The anomalies are characterized by variations of <sup>54</sup>Cr, which is a minor isotope of chromium produced in neutron-rich nuclear statistical equilibrium [11]. Despite years of effort, the possible carrier of these anomalies has not been identified yet. Other rare nuclides may have been co-synthesized with <sup>54</sup>Cr, notably <sup>58</sup>Fe. Depending on the Cr/Fe ratio of the host phase, we may observe correlated variations of <sup>54</sup>Cr and <sup>58</sup>Fe. Iron isotope abundances were measured in Orgueil leaching experiments by TIMS but the precision is limited and no anomalies could be detected [12]. The recent advent of MC-ICP-MS allows the determination of iron isotopic composition with unprecedented precision. Zhu *et al.* [13] measured the iron isotope abundances in macroscopic samples and concluded that iron isotopes were evenly distributed in the solar system. We extend here this study

and analyze iron isotopes in leachate fractions of Orgueil and Allende.

The measurements were performed on a Micromass Isoprobe MC-ICP-MS at the CRPG (Nancy). The background in the iron mass region is high in hard extraction mode due to the presence of argides (ArN, ArO, and ArOH). Introduction of hydrogen in the hexapole collision cell removes the interferences at masses 54 (ArN) and 56 (ArO) but increases that at mass 57 (ArOH). In order to increase the signal/noise ratio at mass 57, we decreased the value of the extraction lens from 40 to 10, which resulted in a large reduction of the noise level from ArOH and prepared solutions ten times more concentrated for compensating for the associated loss in sensitivity. This technique may be applied to abundant elements when the noise level is high. Iron isotopic compositions are reported in  $\epsilon$  units, which are relative deviations in parts per ten thousand.

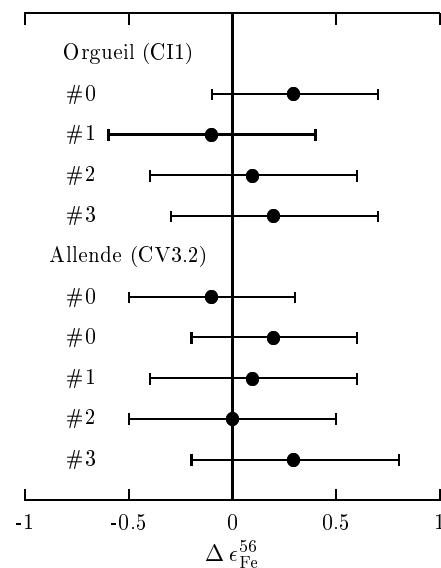


Fig. 1. Iron isotopic composition in Orgueil and Allende leaching fractions,  $\Delta \epsilon_{\text{Fe}}^{56} = \epsilon^{56} - 2/3\epsilon^{57}$ . See [7] for details on the leaching experiments (#0 bulk, #1 CH<sub>3</sub>COOH, #2 HNO<sub>3</sub>, #3 HCl-HF, #4 HNO<sub>3</sub>-HF-HClO<sub>4</sub>). Uncertainties are  $2\sigma$ .

As illustrated in Fig. 1, the intrinsic iron isotopic composition of Orgueil and Allende are homogeneous, which confirms the earlier inference [13] based on

measurements of macroscopic samples that the protosolar nebula was largely homogeneous for iron. The largest variations are expected for  $^{58}\text{Fe}$  and we plan to measure this isotope soon.

**Selenium isotopes:** As in the case of  $^{54}\text{Cr}$ , selenium-82 may have been produced in part during neutron-rich nuclear statistical equilibrium. We may thus expect to observe correlated anomalies between these two nuclides. If the carrier of Mo-m [6, 7], the *s*-enriched molybdenum component, contains some selenium, we may also expect to observe a dichotomy between *pr* and *s* isotopes in leaching experiments. Selenium thus appears to be a promising candidate for identifying isotope anomalies. Such a discovery would be important for tracing accretion of undifferentiated material on differentiated bodies. In the Earth for instance, the selenium content of the mantle is dominated by the late veneer. This may explain why the mantle reservoir is almost unfractionated relative to the chondritic reservoir [14]. If there were isotope anomalies for one of the elements delivered by the late veneer, then we may be able to identify the nature of the material accreted by the Earth after core formation.

Selenium was analyzed on an ICP-MS using a continuous-flow hydride generation system [14]. The sensitivity of this introduction system is very high (300-400 V/ppm) allowing the determination of selenium isotope abundances in a few tens of ng. Argon dimmers form direct isobaric interferences on selenium. They can be eliminated by introducing hydrogen in the collision cell. The main drawback of using hydrogen is that selenium hydrides are formed in the mass spectrometer (3 %), which complicates the data reduction. The ratios were corrected for instrumental and natural mass fractionation by standard bracketing and normalizing the  $^{76}\text{Se}/^{82}\text{Se}$  ratio to a reference value. Selenium hydrides were further corrected by assuming that any variation at mass 77 were the result of hydride interferences. This double normalization technique (for both mass fractionation and hydride interferences) permits the determination of the selenium isotopic composition with high precision. All nuclides are normalized to  $^{82}\text{Se}$ .

As illustrated in Fig. 2, all meteorites measured so far do not deviate from terrestrial composition. As observed for sulfur, the isotopic composition of selenium was probably homogenized in the protosolar nebula. We plan to measure selenium isotopes in leaching fractions using a different separation technique that should prevent variation in the SeH/Se ratio and hence permit the measurement of  $\Delta^{77}\text{Se}$ . We also plan to investigate the mass-dependent variations of the selenium isotopic composition

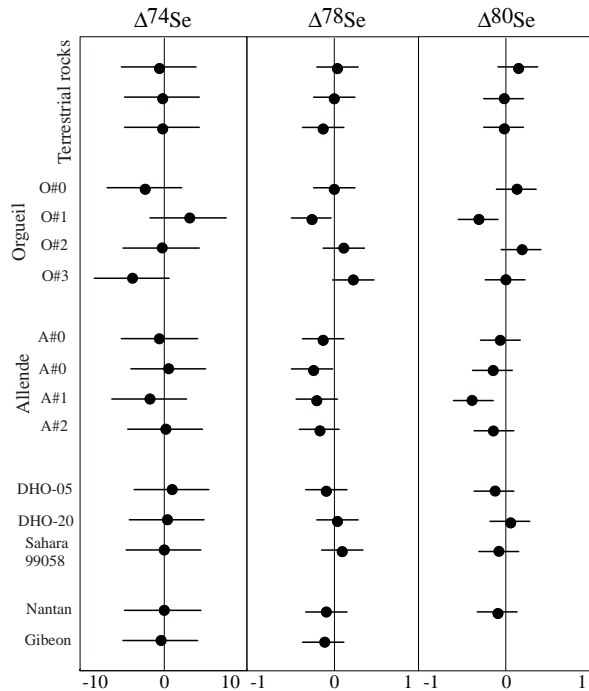


Fig. 2. Selenium isotopic composition. See Fig. 1 caption.  $\Delta^i\text{Se}$  is in permil. Uncertainties are  $2\sigma$ .

**Conclusions:** The isotopic compositions of iron and selenium were homogenized to a large extent in the protosolar nebula. Further work must be done on iron in order to characterize the isotopic composition of  $^{54}\text{Cr}$  rich fractions for  $^{58}\text{Fe}$  with high precision, a work under progress.

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