

NUCLEOSYNTHETIC HETEROGENEITIES OF NEUTRON-RICH ISOTOPES IN CALCIUM ALUMINUM-RICH INCLUSIONS AND BULK SOLAR SYSTEM MATERIALS.

M. Schönbachler¹, W. M. Akram¹, N. H. Williams¹ and I. Leya², ¹School of Earth, Atmospheric and Environmental Sciences, The University of Manchester, Oxford Road, M13 9PL, UK (m.schonbachler@manchester.ac.uk), ²Physical Institute, University of Bern, Switzerland

Introduction: Nucleosynthetic heterogeneities of isotopes in solar system materials can provide important constraints about the dynamic evolution of the protoplanetary disk and the nucleosynthetic sources of solar system materials. Zirconium isotopes represent a powerful tool to address this issue because they have distinct nucleosynthetic sources. While ^{90,91,92,94}Zr are predominantly produced by the *s*-process, the neutron-rich isotope ⁹⁶Zr requires higher neutron fluxes ($> 10^8 \text{ cm}^{-3}$) such that neutron capture can efficiently compete with the β^- decay of ⁹⁵Zr (half-life = 64 days) [1]. Stellar environments that are capable to produce ⁹⁶Zr are supernovae, but also AGB stars synthesize ⁹⁶Zr during short evolutionary phases [2]. Earlier work documented Zr isotope homogeneity in the solar system based on the Zr isotope compositions of various chondrites, eucrites, lunar and terrestrial samples [3]. More recent work at higher precision, however, was able to resolve ⁹⁶Zr heterogeneities between various bulk carbonaceous chondrites and the Earth-Moon system [4]. Similar variations at bulk rock scale for the other neutron-rich isotopes such as ⁵⁰Ti and ⁵⁴Cr are also well established [5-8]. This raises the question, whether Ti, Cr and Zr isotope variations in bulk solar system materials are linked to a single carrier phase, which was heterogeneously distributed within the solar system.

Refractory inclusions: Most calcium aluminum-rich inclusions (CAI) exhibit relative uniform excesses of both $\epsilon^{96}\text{Zr}$ ($\sim +2$) and $\epsilon^{50}\text{Ti}$ ($\sim +9$) [3, 4, 8-10]. Our new Zr isotope data for CAIs further corroborates this uniformity [4]. Only few CAIs display lower $\epsilon^{96}\text{Zr}$ and $\epsilon^{50}\text{Ti}$ values. These inclusions are not petrographically distinct and cover a range of rare earth element patterns similar to the inclusion with excesses. The $\epsilon^{96}\text{Zr}$ and $\epsilon^{50}\text{Ti}$ values of CAIs display a positive correlation [10]. This correlation can be extended to ⁶²Ni and ⁶⁰Fe, although combined Zr, Ni and Ti isotope measurements only exist for the limited dataset of three CAIs [3, 10, 11]. Based on correlated ⁶²Ni and ⁶⁰Fe excesses, Quitté et al. [11] concluded that a neutron-rich stellar event not only delivered ⁶²Ni and ⁹⁶Zr, but also some of the short-lived radionuclides. More combined data is desirable to link nucleosynthetic anomalies to short-lived radionuclides in order to better constrain the timing of the events that lead to the formation of the

solar system. The presence of correlated excesses in the neutron-rich isotopes ⁹⁶Zr, ⁵⁰Ti, and ⁶²Ni in CAIs supports the view that CAIs formed from a reservoir that was well mixed (potentially in a gas phase), but clearly distinct from the rest of the solar system. One possible explanation is that CAIs preserved this distinct heterogeneity because they formed very early before homogenization of the material in the disk had progressed. The original heterogeneity may have been introduced by a late injection of supernova material prior or during the collapse of the solar nebula [12].

Bulk carbonaceous chondrites: Similar to CAIs, bulk carbonaceous chondrites also display correlated excesses of ⁹⁶Zr and ⁵⁰Ti (Fig.1). Mixing calculations show that the ⁹⁶Zr and ⁵⁰Ti excesses in carbonaceous chondrites can be explained by admixing of varying amounts CAIs to average solar system material (Fig. 1). Therefore, CAIs are most likely the carrier phase responsible for the ⁹⁶Zr-⁵⁰Ti variations in bulk carbonaceous chondrites. Trinquier et al. [8] reached the same conclusion based on the $\epsilon^{50}\text{Ti}-\epsilon^{54}\text{Cr}$ relationship in bulk carbonaceous chondrites.

Other bulk meteorites: Other meteorites such as eucrites and ordinary chondrites do not fall on a mixing line between average solar system material and CAIs (Fig. 1). This indicates that CAIs are not responsible for the variable isotopic signatures of these meteorites. Intriguingly, the nucleosynthetic component sampled by CAIs displays a positive ⁹⁶Zr-⁵⁰Ti correlation, while the isotopic compositions of most other analyzed bulk samples (CI and ordinary chondrites, eucrites, lunar and terrestrial samples) show a negative ⁹⁶Zr-⁵⁰Ti correlation, with a slope of -4.33 ± 1.7 (Fig. 1). If this correlation represents a mixing line between two reservoirs with distinct Zr and Ti isotope compositions, the linear correlation indicates that the two reservoirs had similar Zr/Ti ratios. The opposite sign of the two trends (Fig. 1) suggests that CAIs sampled a nucleosynthetic component of distinctively different origin than the remaining samples. The exceptions are carbonaceous chondrites, which appear to be mostly dominated by CAIs. Since carbonaceous chondrites are generally considered to originate from further out in the solar system, this can tentatively be interpreted that a different type of nucleosynthetic material (potentially of supernovae origin) was heterogeneously

distributed in the outer solar system compared to the inner solar system.

Leaching experiments: An often-used approach to identify carrier phases of nucleosynthetic anomalies is to perform leaching experiments on carbonaceous chondrites. This is because many of the original presolar phases have survived in these meteorites. We leached bulk rock samples of Orgueil (CI), Murchison (CM) and Allende (CV) with progressively stronger acids and analyzed them for Zr, Te and Ti isotopes. While Te only shows hints of excesses in the neutron-rich isotope ^{130}Te [13], Zr isotopes reveal a distinct isotopic pattern with high $\epsilon^{96}\text{Zr}$ (up to +50) in the easily leachable fractions (acetic acid), which then decreases to negative $\epsilon^{96}\text{Zr}$ in the later more aggressive leach steps [14]. The most extreme depletion in $\epsilon^{96}\text{Zr}$ (-377.5 ± 3.3) was observed in the last leaching step of Orgueil (Parr bomb dissolution). The extreme ^{96}Zr depletions in the last leaching steps can easily be explained by the presence of presolar SiC grains. However, the ^{96}Zr enrichments in the early leach fractions - particularly for Allende - cannot be due to undissolved SiC grains, because SiC abundances are extremely low in Allende. Moreover, mass balance calculations show that CAIs in Allende can only account for $\epsilon^{96}\text{Zr}$ of +1. The observed ^{96}Zr excesses in the acetic acid leachates of Allende are in the range of +7 to +10 and this indicates the presence of at least one additional unknown phase with anomalous Zr [14].

The Ti isotope data obtained on the same leach fractions depict a different picture. In contrast to bulk rock anomalies, where the ^{50}Ti effects are larger than those in ^{96}Zr (Fig. 1), the leach experiments show the opposite. The ^{50}Ti anomalies are much more subdued, generally showing a trend from negative to positive ^{50}Ti values with increasing acid strength and a maximum $\epsilon^{50}\text{Ti}$ of $+7 \pm 1.6$ for the Parr bomb dissolution step of Murchison. The results are biased because the leachates with the most extreme ^{96}Zr excesses (acetic acid and Parr bomb dissolutions) could often not be analyzed due to low Ti contents. A tentative best-fit line for the Murchison leachates in a $\epsilon^{96}\text{Zr}$ - $\epsilon^{50}\text{Ti}$ four isotope diagram yields a negative slope (-0.23 ± 0.15), which is reminiscent of the negative correlation derived from bulk rock samples (excluding CAIs and most carbonaceous chondrites). The interpretation of the data is complicated because the Zr budget of the leachates is significantly influenced by presolar SiC, whereas mass balance calculation suggests that this is not the case for Ti. The majority of SiC grains (mainstream grains) show ^{50}Ti excesses and ^{96}Zr depletions as measured for the

later leach steps where these refractory grains are dissolved. However, taking mass balance calculations into account, most likely an additional unknown phase (e.g., presolar silicates?) with inversely correlated ^{50}Ti and ^{96}Zr must also be present.

This discussion shows the limitation of leaching experiments to identify the original carrier of bulk rock anomalies. The various nucleosynthetic components and presolar phases resolved in such leaching experiments do not necessarily mirror the phase that is responsible for the observed isotopic anomalies at bulk rock level, because other presolar phases, which are homogeneously distributed throughout the solar system, are also resolved.

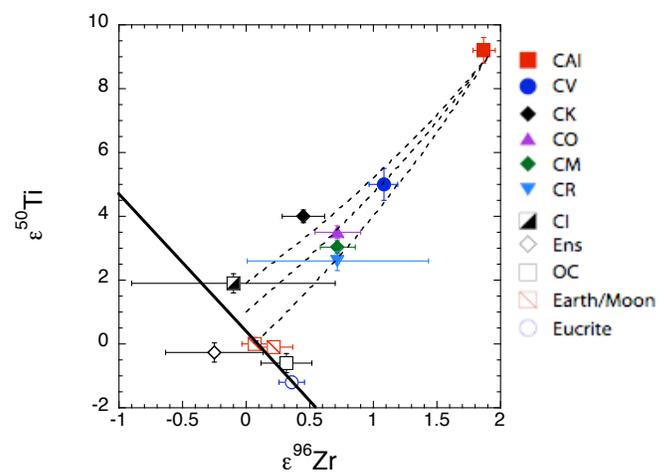


Fig. 1: $\epsilon^{96}\text{Zr}$ versus $\epsilon^{50}\text{Ti}$. Three mixing lines (dashed lines) are shown between CAIs and (i) CI chondrites, (ii) the Earth and (iii) a mixture of both as approximation for average solar system. The black solid line represents the best-fit line defined by CI and ordinary chondrites, eucrites, terrestrial and lunar samples.

References: [1] Nicolussi G., K. et al. (1997) *Science* 277, 1281-1283. [2] Lugaro M. et al. (2003) *Astrophys. J.* 593, 486-508. [3] Schönbachler M. et al. (2003) *EPSL* 216, 467-481. [4] Akram W.M. et al. (2011) *Lunar Planet. Sci.* 42, 1908. [5] Leya I. et al. (2008) *EPSL* 266, 233-244. [6] Qin L. et al. (2010) *GCA* 74, 1122-1145. [7] Trinquier A. et al. (2007) *Astrophys. J.* 655, 1179-1185. [8] Trinquier A. et al. (2009) *Science* 324, 374-376. [9] Harper C.L. et al. (1991) *Lunar Planet. Sci.* 22, 517-518. [10] Leya I. et al. (2009) *Astroph. J.* 702, 1118-1126. [11] Quitté G. et al. (2007) *Astroph. J.* 655, 678-684. [12] Cameron A.G.W. et al. (1977) *Icarus* 30, 447-461. [13] Fehr M.A. et al. (2006) *GCA* 70, 3436-3448. [14] Schönbachler M. et al. (2005) *GCA* 69, 5113-5122.