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The initial abundance and distribution of ^{92}Nb in the Solar System

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ABSTRACT

Niobium-92 is an extinct proton-rich nuclide, which decays to ^{92}Zr with a half-life of 37 Ma. This radionuclide potentially offers a unique opportunity to determine the timescales of early Solar System processes and the site(s) of nucleosynthesis for *p*-nuclei, once its initial abundance and distribution in the Solar System are well established. Here we present internal Nb–Zr isochrons for three basaltic achondrites with known U–Pb ages: the angrite NWA 4590, the eucrite Agoult, and the ungrouped achondrite Ibitira. Our results show that the relative Nb–Zr isochron ages of the three meteorites are consistent with the time intervals obtained from the Pb–Pb chronometer for pyroxene and plagioclase, indicating that ^{92}Nb was homogeneously distributed among their source regions. The Nb–Zr and Pb–Pb data for NWA 4590 yield the most reliable and precise reference point for anchoring the Nb–Zr chronometer to the absolute timescale: an initial ^{92}Nb/^{93}Nb ratio of $(1.4 \pm 0.5) \times 10^{-5}$ at 4557.93 ± 0.36 Ma, which corresponds to a ^{92}Nb/^{93}Nb ratio of $(1.7 \pm 0.6) \times 10^{-5}$ at the time of the Solar System formation. On the basis of this new initial ratio, we demonstrate the capability of the Nb–Zr chronometer to date early Solar System objects including troilite and rutile, such as iron and stony-iron meteorites. Furthermore, we estimate a nucleosynthetic production ratio of ^{92}Nb to the *p*-nucleus ^{92}Mo between 0.0015 and 0.035. This production ratio, together with the solar abundances of other *p*-nuclei with similar masses, can be best explained if these light *p*-nuclei were primarily synthesized by photodisintegration reactions in Type Ia supernovae.

1. Introduction

The proton-rich radionuclide ^{92}Nb decays to ^{92}Zr by electron capture with a half-life of 37 Ma (Holden, 1990). Since Nb and Zr can fractionate from each other during partial melting of the mantle, mineral crystallization and metal–silicate separation (Tiepolo et al., 2001; Wade and Wood, 2001; Klemme et al., 2002), the Nb–Zr system can potentially be used to determine the timescales of silicate differentiation and core segregation for infant planets (Minster and Allègre, 1982). In addition, the initial ^{92}Nb abundance in the Solar System provides constraints on the nucleosynthetic site(s) of *p*-nuclei (*p*- denotes proton-rich) (Harper, 1996; Yin et al., 2000; Dauphas et al., 2003; Meyer, 2003; Hayakawa et al., 2013; Travaglio et al., 2014). These applications require the initial abundance and distribution of ^{92}Nb (expressed as ^{92}Nb/^{93}Nb) in the Solar System to be defined.

Evidence for live ^{92}Nb in the early Solar System was first obtained from the iron meteorite Toluca in which rutile with high Nb/Zr exhibits a ^{92}Zr excess (Harper, 1996). Assuming that the iron meteorite started with initial ^{92}Nb/^{93}Nb and ^{92}Zr/^{90}Zr values identical to those of the chondritic uniform reservoir (CHUR), an interpolation between Toluca rutile and CHUR data was used to estimate the initial ^{92}Nb/^{93}Nb to $(1.6 \pm 0.3) \times 10^{-5}$. Subsequent studies (Sanloup et al., 2000; Yin et al., 2000; Münker et al., 2000) also reported ^{92}Zr variations in meteoritic phases with fractionated Nb/Zr: rutile from the iron meteorite Zagora, zircon from the mesosiderite Chaunskij, and calcium–aluminum rich inclusions (CAIs) from the carbonaceous chondrite Allende. However, the initial ^{92}Nb/^{93}Nb values inferred in the same manner were two orders of magnitude higher ($\sim 10^{-3}$). In contrast, later Zr isotopic studies of zircon from the eucrite Camel Donga (Hirata, 2001) and Allende CAIs (Schönbächler et al., 2003; Akram et al., 2013; Mane et al., 2014) did not reveal ^{92}Zr variations due to ^{92}Nb decay, constraining the initial ^{92}Nb/^{93}Nb of the Solar System to $<10^{-4}$. Moreover, Schönbächler et al. (2002) determined internal Nb–Zr isochrons for the ordinary (H6) chondrite Estacado and the

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mesosiderite Vaca Muerta, and obtained initial $^{92}Nb/^{93}Nb$ values of $\sim 10^{-5}$.

There are several possible causes of the extensive variation in the estimated initial $^{92}Nb/^{93}Nb$ ratios (10^{-3} to 10^{-5}): (1) significant differences in the Nb–Zr closure age (>200 Ma) among the studied meteorites, (2) heterogeneous distribution of ^{92}Nb in the early Solar System, (3) highly variable initial Zr isotope compositions among the studied meteorites and CHUR, and (4) analytical artifacts. To evaluate these possibilities and firmly establish the initial abundance and distribution of ^{92}Nb in the Solar System, it is essential to define internal isochrons for multiple meteorites that originate from distinct parent bodies and whose absolute ages are precisely known. For instance, scenario (2) or (3) renders the Nb–Zr isochron regressions using CHUR and non-chondritic materials invalid. Schönbächler et al. (2002) utilized the internal isochron approach, yet the analyzed meteorites include components of different origins and their formation ages are uncertain, which prohibits a precise determination of the solar initial ^{92}Nb abundance.

Here we present internal Nb–Zr isochrons of three unbrecciated achondrites with known U–Pb ages (Amelin et al., 2011a; Iizuka et al., 2013, 2014, 2015a): the angrite NWA 4590, the eucrite Agoult and the ungrouped achondrite Ibitira. The internal isochrons allow us to precisely determine the initial abundance of ^{92}Nb in the Solar System and to assess its distribution in the solar nebula. We will discuss the implications of the results for early Solar System chronology and the origin of *p*-nuclei.

2. Samples

Northwest Africa 4590 is a very fresh angrite with plutonic igneous textures. It mainly comprises clinopyroxene, anorthite, olivine, kirschsteinite, ulvöspinel, and accessory phosphates and troilite (Kuehner and Irving, 2007). The Re–Os and highly siderophile element geochemistry indicate that while several angrites provide evidence for contamination of exogenous chondritic materials to their protoliths, NWA 4590 is the least-contaminated angrite (Riches et al., 2012). The Pb–Pb age of 4557.93 ± 0.36 Ma, which dates crystallization, was obtained from acid-leached pyroxene using the $^{238}U/^{235}U$ value of 137.789 ± 0.021 measured for the whole rock sample (Amelin et al., 2011a). The Pb–Pb isochron age is consistent with the Mn–Cr and Hf–W isochron ages of 4557.1 ± 1.0 Ma (Yin et al., 2009) and 4557.8 ± 0.6 Ma (Kleine et al., 2009), which are calculated using the Pb–Pb isochron age of 4563.37 ± 0.25 Ma for the angrite D'Orbigny as a time anchor (all Mn–Cr and Hf–W ages in this paper are recalculated using Mn–Cr data of Glavin et al., 2004, Hf–W data of Kleine et al., 2012 and the U–Pb age of Brennecka and Wadhwa, 2012 for D'Orbigny). In addition, the phosphate Pb–Pb age (4557.381 ± 0.066 Ma; Amelin et al., 2011a) is slightly younger than that of pyroxene and constrains the cooling rate of the parent rock to 590 ± 240 K/Ma. Abundances of trace elements including Nb and Zr were previously determined by laser ablation-inductively coupled plasma mass spectrometry (LA-ICPMS) for the constitute minerals (Amelin et al., 2011b). The results demonstrate that the principal carrier phases of Zr are ulvöspinel and pyroxene, and that the former has significantly higher Nb/Zr than the latter.

Agoult is an unbrecciated, fine-grained granulitic eucrite with no chemical evidence for weathering (Yamaguchi et al., 2009). It has remnant subophitic textures composed of anhedral pyroxene and elongated plagioclase with abundant ca. 120 °C triple-junction, which indicates significant recrystallization of a basaltic protolith during high-grade metamorphism (Yamaguchi et al., 2009). Minor phases are opaque minerals including ilmenite and Ti-chromite, silica mineral, phosphates, troilite, zircon and baddeleyite (Yamaguchi et al., 2009). Importantly, some zircon grains in Agoult are exceptionally large (up to ~80 μm) for eucrites, allowing us to perform the first combined high-precision U–Pb and Zr isotope analyses of single meteorite zircon grains. The Agoult zircon is typically accompanied by ilmenite and tridymite, while ilmenite often includes needles of baddeleyite (Iizuka et al., 2015a). Titanium contents in these zircon grains indicate that they crystallized at subsolidus temperatures of ~900 °C (Iizuka et al., 2015a). The mineral assemblage and sub-solidus crystallization temperatures provide evidence that the Agoult zircon formed through ZrO_2 release (baddeleyite exsolution) from ilmenite followed by reaction with the surrounding silica during high-temperature metamorphism. The zircon crystallization age of 4554.5 ± 2.0 Ma was determined from eight grains with concordant U–Pb systems (Iizuka et al., 2015a), three of which were analyzed for Zr isotopes in this study. However, substantially younger dates were obtained from plagioclase and pyroxene fractions of Agoult (Iizuka et al., 2013): the plagioclase fractions gave a Pb–Pb isochron age of 4532.2 ± 1.0 Ma, whereas the pyroxene fractions yielded scattered data with Pb–Pb model ages from 4529 to 4523 Ma. All of the zircon, plagioclase and pyroxene Pb–Pb ages were calculated using the whole rock $^{238}U/^{235}U$ value of 137.709 ± 0.016 (Iizuka et al., 2013). These observations indicate that Agoult underwent two metamorphic events at 4554 Ma and ca. 4530 Ma and that the latter event reset the U–Pb system in plagioclase and pyroxene, but not in zircon.

Ibitira is a fine-grained, unbrecciated vesicular basaltic achondrite. It is mainly composed of clinopyroxene (~60%) that shows coarse exsolution lamellae of augite from pigeonite and plagioclase (~30%). It also features minor tridymite, intergrowths of ilmenite and Ti-chromite, kamacite, troilite, phosphate and olivine (Wilkening and Anders, 1975; Steele and Smith, 1976). Ibitira has long been classified as an eucrite based on the mineral assemblage, but recent detailed geochemical and O isotopic studies reveal that the parent body of Ibitira is distinct from that of the eucrites (Wiechert et al., 2004; Mittlefehldt, 2005; Scott et al., 2009). The U–Pb dating of acid-leached pyroxene and whole rock fractions, together with whole rock $^{238}U/^{235}U$ measurements, yielded a Pb–Pb isochron age of 4556.75 ± 0.57 Ma, which was interpreted as the timing of the prolonged thermal metamorphism associated with the pyroxene exsolution and chemical equilibration (Iizuka et al., 2014). The Pb–Pb age is in good agreement with the Mn–Cr isochron ages of 4557.4 ± 2.5 Ma (Lugmair and Shukolyukov, 1998) and 4555.9 ± 3.2 Ma (Yin et al., 2009) defined by the whole rock, silicate minerals (essentially pyroxene) and chromite, calculated using the angrite D'Orbigny time anchor (Glavin et al., 2004; Brennecka and Wadhwa, 2012). Moreover, these ages are consistent with the ^{146}Sm–^{142}Nd and I–Xe ages (Prinzhofer et al., 1992; Claydon et al., 2013), even though the ^{147}Sm–^{143}Nd systematics suggests partial Sm–Nd re-equilibration between plagioclase and phosphate (Prinzhofer et al., 1992). This likely occurred during a moderate shock event dated at 4.49 Ga by Ar–Ar chronology (Bogard and Garrison, 2003).

3. Methods

Three different analytical schemes were used in this study (Table 1). Analytical schemes A and B were used for fractions of pyroxene, spinel, opaque minerals, and whole rock, while scheme C was applied to zircon. The mineral fractions were hand-picked from the disaggregated specimens of NWA 4590, Agoult and Ibitira, whereas the whole rock fractions represent disaggregated materials without mineral separation. All the mineral and whole rock fractions were cleaned with distilled acetone and digested with a concentrated $HF–HNO_3$ mixture. The spinel and opaque mineral fractions were digested using a 125 ml Parr® bomb. The digested fractions were converted to a soluble form by repeated evaporation with concentrated HNO_3, and then dissolved in 6 M HCl.