



Abundance, distribution, and origin of ^{60}Fe in the solar protoplanetary disk

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ABSTRACT

Meteorites contain relict decay products of short-lived radionuclides that were present in the protoplanetary disk when asteroids and planets formed. Several studies reported a high abundance of ^{60}Fe ($t_{1/2}=2.62 \pm 0.04$ Myr) in chondrites ($^{60}\text{Fe}/^{56}\text{Fe} \sim 6 \times 10^{-7}$), suggesting that planetary materials incorporated fresh products of stellar nucleosynthesis ejected by one or several massive stars that exploded in the vicinity of the newborn Sun. We measured $^{58}\text{Fe}/^{54}\text{Fe}$ and $^{60}\text{Ni}/^{58}\text{Ni}$ isotope ratios in whole rocks and constituents of differentiated achondrites (ureilites, aubrites, HEDs, and angrites), unequilibrated ordinary chondrites Semarkona (LL3.0) and NWA 5717 (ungrouped petrologic type 3.05), metal-rich carbonaceous chondrite Gujba (CBa), and several other meteorites (CV, EL H, LL chondrites; IIIAB, IVA, IVB iron meteorites). We derive from these measurements a much lower initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio of $(11.5 \pm 2.6) \times 10^{-9}$ and conclude that ^{60}Fe was homogeneously distributed among planetary bodies. This low ratio is consistent with derivation of ^{60}Fe from galactic background ($^{60}\text{Fe}/^{56}\text{Fe} \approx 2.8 \times 10^{-7}$ in the interstellar medium from γ -ray observations) and can be reconciled with high $^{26}\text{Al}/^{27}\text{Al} \sim 5 \times 10^{-5}$ in chondrites if solar material was contaminated through winds by outer layers of one or several massive stars (e.g., a Wolf–Rayet star) rich in ^{26}Al and poor in ^{60}Fe . We present the first chronological application of the ^{60}Fe – ^{60}Ni decay system to establish the time of core formation on Vesta at $3.7^{+2.5}_{-1.7}$ Myr after condensation of calcium–aluminum–rich inclusions (CAIs).

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1. Introduction

Extinct radionuclides have proven to be extremely useful for studying the astrophysical context of solar system formation, the relative chronology of early solar system events, the irradiation history of early formed solids, the timescale of nucleosynthetic processes, and the thermal history of planetary bodies (see recent reviews by Meyer and Clayton, 2000; McKeegan and Davis, 2004; Wadhwa et al., 2006; Wasserburg et al., 2006; Dauphas and Chaussidon, 2011). Many of these extinct radionuclides originated from the long-term chemical evolution of the Galaxy (e.g., ^{92}Nb , ^{129}I , ^{146}Sm , and ^{244}Pu ; Clayton, 1985, 1988; Nittler and Dauphas, 2006; Huss et al., 2009) or may have been produced by particle irradiation around the young Sun (e.g. ^{10}Be and ^{36}Cl , McKeegan et al., 2000; Hsu et al., 2006; Jacobsen et al., 2011). Others such as ^{26}Al , whose abundances exceed expectations for inheritance from galactic background (Meyer and Clayton, 2000; Wasserburg et al., 2006; Huss et al., 2009) and production by particle irradiation (Marhas et al., 2002; Duprat and Tatischeff, 2007), were most likely injected by a nearby stellar source.

Iron-60 ($t_{1/2}=2.62$ Myr; Rugel et al., 2009) has been the focus of much work recently because it cannot be produced by particle

irradiation in the solar protoplanetary disk (Lee et al., 1998) and a high $^{60}\text{Fe}/^{56}\text{Fe}$ ratio in meteorites ($> 3 \times 10^{-7}$) would tie the birth of the solar system to the explosion of a nearby supernova (Wasserburg et al., 1998). However, the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio at the time of solar system formation is highly uncertain with estimates that span over two orders of magnitude (Birck and Lugmair, 1988; Shukolyukov and Lugmair, 1993a,b; Tachibana et al., 2003, 2006; Mostefaoui et al., 2004; 2005; Sugiura et al., 2006; Quitté et al. 2007, 2010, 2011; Guan et al., 2007; Tang and Dauphas, 2011a, 2011b, 2012; Spivak-Birndorf et al., 2011, 2012; Moynier et al., 2011; Marhas and Mishra, 2012; Mishra and Chaussidon, 2012; Telus et al., 2012). Time zero in early solar system chronology is marked by calcium–aluminum–rich inclusions (CAIs), which are the oldest solar system solids. In CAIs, the only significant carriers of Fe and Ni are opaque assemblages (previously known as Fremdlinge; Palme and Wlotzka 1976; Wark and Lovering 1976; El Goresy et al., 1978; Blum et al., 1988; Sylvester et al., 1990) and smaller sub-micron refractory metal nuggets (Blander et al., 1980; Wark 1986; Berg et al., 2009), which have low Fe/Ni ratios and are therefore not very well suited to constrain the $^{60}\text{Fe}/^{56}\text{Fe}$ ratio. Another complication is the fact that CAIs display isotopic anomalies for ^{62}Ni that correlate with ^{60}Ni , so it is difficult to ascribe ^{60}Ni isotope variations solely to decay of ^{60}Fe (Birck and Lugmair, 1988; Quitté et al., 2007). For these reasons, Birck and Lugmair (1988) and Quitté et al. (2007) were only able to establish an upper-limit on the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio of less than $\sim 1.6 \times 10^{-6}$.

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The first evidence for ^{60}Fe in solar system material was found by thermal ionization mass spectrometry (TIMS) in the form of excess ^{60}Ni in mineral separates from eucrites (Shukolyukov and Lugmair, 1993a,b), a group of basaltic meteorites that are thought to come from the crust of asteroid Vesta (McCord et al., 1970; De Sanctis et al., 2012; Russell et al., 2012). Internal isochrons in the eucrites Chervony Kut and Juvinas yielded initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratios of 3.9×10^{-9} and 4.3×10^{-10} , respectively. However, the closure time for the ^{60}Fe – ^{60}Ni system in these meteorites was unknown, hindering a reliable estimate of the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio at solar system birth (Mostefaoui et al., 2004). Subsequent studies have yielded widely variable initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio estimates.

In situ measurements by secondary ion mass spectrometry (SIMS) of constituents of unequilibrated ordinary chondrites, such as pyroxenes in chondrules, have given elevated $^{60}\text{Fe}/^{56}\text{Fe}$ initial ratios at CAI formation of around $\sim 6 \times 10^{-7}$ (Tachibana et al., 2003, 2006; Mostefaoui et al., 2004, 2005; Guan et al., 2007; Marhas and Mishra, 2012; Mishra et al., 2010; Mishra and Chaussidon, 2012). Ogliore et al. (2011) identified a statistical bias that affected some SIMS measurements (Tachibana et al., 2003, 2006), leading Telus et al. (2012) to revise the initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio to 5×10^{-8} or higher. More recent studies have also addressed these analytical issues and have found a high initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio of $\sim 6 \times 10^{-7}$ at solar system formation (Marhas and Mishra, 2012; Mishra and Chaussidon, 2012).

Measurements of differentiated achondrites (meteorites that were once molten) by various methods have given much lower ratios of $\sim 2 \times 10^{-8}$ (Sugiura et al., 2006; Quitté et al., 2010, 2011; Tang and Dauphas, 2011a, 2011b, 2012; Spivak-Birndorf et al., 2011, 2012). The reason for this discrepancy is unknown but some have argued that this could be due to a heterogeneous distribution of ^{60}Fe (Sugiura et al., 2006; Quitté et al., 2010). As discussed by Wasserburg et al. (1998), solving the issue of the initial abundance of ^{60}Fe and of its distribution in early solar system materials is critical to assess the plausibility of the scenario of supernova-triggered solar system formation (Cameron and Truran, 1977; Boss and Keiser, 2012).

To study the initial abundance of ^{60}Fe and its distribution in the disk, we have measured the Ni isotopic compositions of bulk HEDs, angrites and mineral separates from quenched angrite D'Orbigny. Motivated by the large discrepancy between achondrite and chondrite measurements, we have also studied the Ni isotopic compositions of whole rocks, chondrules and mineral separates from CBa (Bencubbin-type) chondrite Gujba, as well as two ordinary chondrites Semarkona (LL group) and NWA 5717 (ungrouped), which have experienced minimal thermal metamorphism (petrologic types 3.0 and 3.05, respectively, on a scale that starts at 3.0 and extends to 6; Grossman and Brearley, 2005). Our results demonstrate that the $^{60}\text{Fe}/^{56}\text{Fe}$ initial ratio in the solar protoplanetary disk was $(11.5 \pm 2.6) \times 10^{-9}$ and that ^{60}Fe was homogeneously distributed among large planetary objects.

2. Methodology

Details of the methodology are available in Appendix B.

2.1. Sample preparation and digestion

Bulk achondrites (8 angrites, 9 HEDs, 2 ureillites, and 1 aubrite) weighing up to 1 g were first polished with abrasive paper and rinsed with acetone to get rid of fusion crust and other surface contaminants. D'Orbigny and NWA 5717 were fragmented by high voltage pulse power fragmentation (Selfrag) for mineral separation. The fragmented samples were processed with a hand magnet, sieves and sodium polytungstate solution. Using these

procedures, metal, multiple silicate grain sizes and samples with different densities (below or above 3.10 g/cm^3) were separated. Bulk chondrules from Semarkona and NWA 5717 were hand-picked for analyses. Six NWA 5717 entire chondrules and three silicate size fractions (100–166 μm , 166–200 μm and $> 200 \mu\text{m}$) were washed before powdering in 1 M HCl for 30 min in order to remove surface-sited metal, sulfide, and other soluble phases. Twenty-one chondrules from CBa chondrite Gujba (a meteorite fall with little evidence of terrestrial alteration) were sampled using a New Wave Research Micromill apparatus. Silicate chondrules were crushed and the fragments with adhering metal sensitive to a hand magnet were removed before digestion. Whole rocks of several chondrites (1 CV, 1 CM, 1 EL, 1 EH, and 1 LL) and iron meteorites (1 IIAB, 1 IIIAB, 1 IVA, and 2 IVB) were also analyzed.

Iron meteorites and metal chondrules from Gujba were digested in aqua regia. All other samples were crushed in an agate mortar, and the sample powder was dissolved in a 5–30 mL mixture of concentrated HF–HNO₃ (in a 2:1 ratio by volume) in a Teflon beaker placed on a hot plate at $\sim 90^\circ\text{C}$ for 5–10 days. The solution was subsequently evaporated to dryness and redissolved in a 5–30 mL mixture of concentrated HCl–HNO₃ (2:1 ratio). The solutions were dried down and taken back to solution with a minimum amount of concentrated HCl ($\sim 11 \text{ M}$) for loading on the first column.

2.2. Protocols for Ni and Fe purification

Chemical purification of Ni was achieved in a three-step procedure.

- U/TEVA Cartridge.* The 2 mL U/Teva cartridges (length=2.7 cm, diameter=0.8 cm) were conditioned with 10 mL of concentrated HCl ($\sim 11 \text{ M}$). Approximately 3/4 of the sample solution in concentrated HCl was loaded onto the cartridge; the rest was kept for Fe/Ni determination, other analyses, and as a safety aliquot. The load solution was collected in clean Teflon beakers and an additional 10 mL of concentrated HCl was passed through the resin and collected in the same beaker. This eluate contained Ni together with other matrix elements (Horwitz et al., 1992).
- Cation exchange.* The Ni elution cut from the first column chemistry was evaporated and re-dissolved in 5 mL of a mixture of 20% 10 M HCl–80% acetone (by volume) and loaded onto 5 ml pre-cleaned Bio-Rad AG50-X12 200–400 mesh hydrogen-form resin in teflon columns (length=40 cm, diameter=0.4 cm), previously conditioned with 10 mL 20% 10 M HCl–80% acetone. After loading the sample solution and rinsing with 30 mL 20% 10 M HCl–80% acetone mixture, Ni was collected by eluting 150 mL of the HCl–acetone mixture (Strelow et al., 1971). This HCl–acetone column was repeated 5 times to ensure thorough separation of Mg from Ni.
- Anion exchange.* Zinc was removed using a third column filled with 1 mL Bio-Rad AG1W-X8 anionic ion exchange resin (length=2 cm, diameter=0.8 cm) in 8 M HBr medium (Moynier et al., 2006).

Nickel blanks for the entire procedure ranged from 20 to 35 ng, which is insignificant relative to the amounts of Ni in the samples. Nickel yields for each step were close to 100% and the overall yield of the procedure was 85–100% except for Ibitira (around 60%; Table S1). Achondrites and chondrules have very low Ni contents (e.g., 0.5 μg for some HED meteorites) and the isotopic abundance of ^{64}Ni could not be reliably measured in these samples but we were able to measure ^{64}Ni in bulk chondrites

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