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THE ELUSIVE ^{60}Fe IN THE SOLAR NEBULA

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ABSTRACT

No ^{60}Ni or ^{61}Ni anomalies have been detected in troilite inclusions from Muonionalusta, a 4565.3 ± 0.1 Ma old IVA iron meteorite, to the level of the analytical precision of 10 ppm. Because Muonionalusta troilite is very old, has a high Fe/Ni ratio, and is free of ^{61}Ni anomalies, it is the ideal material in which to search for potential excesses of ^{60}Ni produced by the decay of ^{60}Fe ($t_{1/2} = 2.62$ Ma). The ^{60}Ni/^{58}Ni and Fe/Ni ratios (up to 1680) measured here imply an upper limit for the initial ^{60}Fe/^{56}Fe for the Muonionalusta troilite of 3×10^{-9}. Assuming that ^{60}Fe was homogeneously distributed across the nebula, this result suggests that the solar system initial ^{60}Fe/^{56}Fe ratio was less than 5×10^{-9}, which is lower than previously measured by at least a factor of 50.

Key words: astrochemistry – minor planets, asteroids: general – nuclear reactions, nucleosynthesis, abundances – protoplanetary disks

Online-only material: color figure

1. INTRODUCTION

The solar system is a mixture of components from many astrophysical sources with different chemical and isotopic compositions. In general, mixing of the different presolar components in the nebula was efficient enough for the isotope compositions of planetary material to become remarkably homogeneous. For some time, however, it has been recognized that, in the inner solar system, mixing was incomplete, whether considering isotopes or the elements themselves (Birck 2004). Since the 1970s, isotopic anomalies have been observed at the mineral scale for many elements, particularly in certain types of Ca–Al-rich inclusions (CAIs) from the Allende meteorite known as fractionated and unknown nuclear effect inclusions (Wasserburg 1977; Clayton 1978). Although a chemical origin has also been proposed (Fujii et al. 2006), these mineral-scale anomalies are usually ascribed to incomplete mixing of the products of stellar nucleosynthesis in the early solar system (Birck 2004). Recent improvements in analytical methods and instrumentation have permitted resolution of progressively smaller isotopic anomalies for a number of elements (Ca, Cr, Ti, Ni, Mo, Ru, Ba, Sm, Nd, W) in bulk meteorites (Simon et al. 2010; Dauphas et al. 2004, 2002; Yin et al. 2002; Hidaka et al. 2003; Podosek et al. 1997; Trinquier et al. 2007; Chen et al. 2003; Papanastassiou et al. 2004; Leya et al. 2008; Ranen & Jacobsen 2006; Carlson et al. 2007; Bizzarro et al. 2007; Andreasen & Sharma 2006; Moynier et al. 2010).

The study of decay products of short-lived ($t_{1/2} \leqslant 100$ Ma) radionuclides in the solar nebula provides important clues about the stellar environment and the timescale of events that occurred in the early solar system. Short-lived radionuclides can be synthesized by either particle irradiation from the energetic early Sun or stellar nucleosynthesis (Lee et al. 1998). The origin and interpretation of the presence of extinct radionuclides in meteorites are still open for discussion (Gounelle et al. 2001; Desch et al. 2004; Russell & Gounelle 2001). In this context, ^{60}Fe ($t_{1/2} = 2.62$ Ma) is particularly important because it can only be synthesized efficiently by stellar nucleosynthesis (Tur et al. 2010; Limongi & Chieffi 2006; Timmes et al. 1995). Its presence in meteorites hence would provide unambiguous evidence for the arrival of debris from either a supernova explosion or an asymptotic giant branch (AGB) star in the neighborhood of the nascent Sun 15 Ma or less before its formation (Cameron & Truran 1977; Meyer & Clayton 1999; Wasserburg et al. 2006). The explosion of a supernova could have been the very event that triggered the formation of the solar system (Cameron & Truran 1977; Vanhala & Boss 2000). However, cosmic rays also can lead to the production of limited quantities of ^{60}Fe in meteorites. Thus, in any discussion of "excess" ^{60}Fe it is important to either rule out or correct for cosmic-ray effects.

Fossil ^{60}Fe is detected in meteorites from the correlation of excesses of the daughter ^{60}Ni with the corresponding Fe/Ni ratios. The first hint of ^{60}Ni excess was discovered by Birck & Lugmair (1988) in a refractory inclusion from the Allende meteorite but the limited data set left the origin of this anomaly uncertain. Further indications of fossil ^{60}Fe were found in the basaltic eucrites Chervony Kut, with an initial ^{60}Fe/^{56}Fe ratio of 7×10^{-9} (Shukoliukov & Lugmair 1993a), and Juvinas, with an initial ^{60}Fe/^{56}Fe ratio of 4×10^{-10}(Shukoliukov & Lugmair 1993b). Tachibana et al. (2006) found a correlation between ^{60}Ni and Fe/Ni in FeO-rich pyroxene chondrules in Semarkona and Bishumpur and calculated an ^{60}Fe/^{56}Fe ratio of (2.2–3.7) × 10^{-7}. So far, however, no internal isochron has been established to support isotopic homogenization of iron at a given moment during nebular evolution, and the inferred initial ^{60}Fe/^{56}Fe value remains unresolved.

Because troilite (FeS) generally is characterized by high Fe/Ni ratios (⩾1000), the inferred ^{60}Fe/^{56}Fe ratio does not depend on the slope of a particular isochron and troilite, and, therefore, seems particularly suitable as material in which to search for fossil ^{60}Fe. So far, two large ^{60}Ni anomalies have been measured in troilite from the primitive meteorite Semarkona, an LL 3.0 ordinary chondrite. These anomalies imply an initial

$^{60}Fe/^{56}Fe$ ratio of $(0.92 \pm 0.24) \times 10^{-6}$ (Mostefaoui et al. 2005). Quitté et al. (2006) reported only small ^{60}Ni deficits and ^{61}Ni excesses in troilite from a range of different iron meteorites and suggested they had formed after all the ^{60}Fe had decayed, i.e., $\geqslant$10 Ma after solar system formation, and that they reflect nucleosynthetic anomalies. Cook et al. (2008) analyzed other troilite inclusions from more iron meteorites and confirmed the deficits observed by Quitté et al. (2006) for ^{60}Ni. In contrast, Chen et al. (2009) reported that troilite from iron meteorites and ordinary chondrites are free of Ni isotope anomalies.

The search for ^{60}Fe in the early solar system is expected to be most successful in early formed material with high Fe/Ni ratios. The parent body of angrites is 4564.42 ± 0.12 Ma old (Amelin 2008) and Quitté et al. (2010) determined that its initial $^{60}Fe/^{56}Fe$ ratio is $\approx(3.1 \pm 0.8) \times 10^{-9}$. Although tungsten isotope evidence (Schersten et al. 2006; Qin et al. 2008) indicates that most iron meteorites are formed within a few Ma of CAIs, they show no evidence of having ^{60}Ni excesses (Markowski et al. 2006). The exact ages of these samples, however, remain poorly determined. Recently, troilite from Muonionalusta (an IVA iron meteorite) turned up a strikingly old precise absolute Pb–Pb age (4565.3 ± 0.1 Ma; Blichert-Toft et al. 2010a), which, after allowing for a correction for cooling of 1–2 Ma, indicates that the IVA parent body separated within 1 Ma of the formation of CAIs (Blichert-Toft et al. 2010a; Bouvier et al. 2007; Jacobsen et al. 2008). In addition, Muonionalusta, selected for this study because of its old and precisely known age, is highly depleted in Cu, which is an important feature because Cook et al. (2008) argued that cosmic-ray interactions with Cu unrelated to supernovae have the potential of producing measurable effects on ^{60}Ni and ^{61}Ni abundances. We therefore expect interference effects due to Cu to be minimal for troilite from Muonionalusta. In order to investigate the presence of ^{60}Fe in the early solar system, we analyzed the Ni isotopic composition of the same Muononialusta troilite solutions as used by Blichert-Toft et al. (2010a) for Pb–Pb chronology. We also report Ni isotopic compositions for troilite from Nantan (group IAB), an iron meteorite known to be well shielded from cosmogenic effects (Nishiizumi et al. 2005), and Mundrabilla (group IAB), a shocked iron meteorite for which large deficits of ^{60}Ni and ^{61}Ni were previously reported by Cook et al. (2008).

2. SAMPLES AND ANALYTICAL PROCEDURES

The two troilite inclusions (ML1 and ML2) from Muonionalusta selected for this study are described in detail in Blichert-Toft et al. (2010a), while those from Nantan and Mundrabilla are described in equal amount of detail in Blichert-Toft et al. (2010b). The removal of the troilite inclusions from their enclosing host iron, sample preparation, sequential leaching techniques, dissolution procedures, and the extraction of Fe, all of which pertain to the troilite inclusions analyzed in the present work, are likewise fully documented in Blichert-Toft et al. (2010a, 2010b). We used pliers to cut off most of the iron and a press to bend the whole sample such as to pop out the troilite inclusion from the remaining iron. Whenever the sample was in contact with a stainless steel tool (hammer, press, or chisel), the interface was protected with thick plastic or aluminum foil. Therefore, the troilites have not been contaminated by stainless steel during sample preparation. Aliquots for Ni isotope analysis were taken from the troilite sample solutions prepared at the Ecole Normale Supérieure in Lyon after Fe had been extracted and prior to any of the column chromatography purification

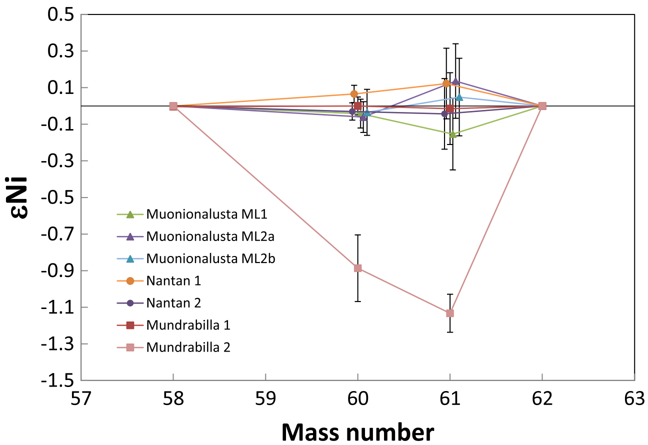

Figure 1. ϵNi in troilite from iron meteorites. The data are normalized to $^{62}Ni/^{58}Ni$ of 0.05339 using the exponential law (Marechal et al. 1999). The errors are reported as 2 standard error (2se) of the replicated measurements. All of the troilite inclusions (but one) analyzed in this study were found to have Ni isotopic compositions similar to the terrestrial standard within the level of measurement precision of 10 ppm. One troilite sample from Mundrabilla shows an ≈ -1 ϵ-unit deficit in both ^{60}Ni and ^{61}Ni.

(A color version of this figure is available in the online journal.)

steps used for the Pb–Pb isotope work of Blichert-Toft et al. (2010a, 2010b). Nickel was then separated from these aliquots following the procedure described in Moynier et al. (2007). The Ni isotopic compositions were measured on a Thermo Scientific Neptune multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at Washington University in St. Louis. The purified Ni solutions (3 ppm in 0.05 HNO_3) were introduced into the mass spectrometer using a 100 μml minute^{-1} PFA nebulizer and cyclonic spray chamber. The measurements were done in high-resolution mode on the peak shoulder in order to resolve the isobaric interference of $^{40}Ar^{18}O$ with ^{58}Ni. The intensities of masses 57, 58, 60, 61, 62, and 64 were measured on the Faraday cups L3, L2, central, H1, H2, and H4, respectively.

3. RESULTS

The Ni isotope data are given in Table 1 and shown in Figure 1 as epsilon units (deviation in parts per 10,000 relative to the composition of the in-house Ni standard) after internal normalization to $^{62}Ni/^{58}Ni$ of 0.05339 and using an exponential law (Marechal et al. 1999). The errors are reported as 2 standard error (2se) of the replicated measurements. All of the troilite samples (but one) analyzed in this study were found to have Ni isotope compositions similar to the terrestrial standard within the level of analytical precision of 10 ppm. The one exception is one of the troilite inclusions from Mundrabilla, which shows an $\approx$1 ϵ-unit deficit in both ^{60}Ni and ^{61}Ni.

4. DISCUSSION

The absence of resolvable ^{61}Ni anomalies from the 4565.3 Ma old troilite of Muonionalusta is additional evidence that this meteorite is a particularly suitable sample for assessing the initial $^{60}Fe/^{56}Fe$ ratio of the solar system. From the Fe/Ni ratio and the error on the $\epsilon^{60}Ni$ of the Muonionalusta sample ML2b, we calculate an upper limit for the $^{60}Fe/^{56}Fe$ ratio at the time of formation of these sulfides to be $\leqslant 3 \times 10^{-9}$. Assuming homogeneous distribution of ^{60}Fe within the early solar system (Dauphas et al. 2008; Moynier et al. 2009), an upper limit for its initial $^{60}Fe/^{56}Fe$ ratio is 5×10^{-9}.

Table 1
Ni Isotopic Compositions of Six Different Troilite Inclusions from Three Different Iron Meteorites (See the Text for Definition of the ϵ Notation)

Sample Name	Type	δ^{60}Ni	δ^{61}Ni	δ^{62}Ni	ϵ^{60}Ni	ϵ^{61}Ni	^{56}Fe/^{58}Ni	n[a]
Muonionalusta ML1	troilite IVA	1.05 ± 0.02	1.56 ± 0.03	2.08 ± 0.04	−0.04 ± 0.08	−0.15 ± 0.20	50	8
Muonionalusta ML2a	troilite IVA	0.92 ± 0.08	1.39 ± 0.12	1.82 ± 0.16	−0.06 ± 0.08	0.14 ± 0.20	80	7
Muonionalusta ML2b	troilite IVA	0.73 ± 0.33	1.10 ± 0.50	1.44 ± 0.65	−0.01 ± 0.10	0.13 ± 0.16	1680	3
Nantan 1	troilite IAB	0.28 ± 0.05	0.41 ± 0.07	0.54 ± 0.09	0.07 ± 0.04	0.12 ± 0.20	...	8
Nantan 2	troilite IAB	1.26 ± 0.04	1.87 ± 0.05	2.48 ± 0.08	−0.03 ± 0.06	−0.04 ± 0.14	...	9
Mundrabilla 1	troilite IAB	−0.95 ± 0.20	−1.41 ± 0.28	−1.86 ± 0.38	0.00 ± 0.05	−0.01 ± 0.20	...	5
Mundrabilla 2	troilite IAB	−1.55 ± 0.11	−2.29 ± 0.13	−2.87 ± 0.18	−0.89 ± 0.18	−1.13 ± 0.10	...	3

Notes. ^{x}Ni/^{58}Ni normalized to ^{62}Ni/^{58}Ni = 0.05339. The superscript $x = 60$ or 61. δ^{x}Ni is the stable isotope fractionation in permil notation as referenced to the standard Spex CL1-130Ni.
[a] Number of sample measurements.

The present upper limit for the ^{60}Fe/^{56}Fe ratio of the Muonionalusta troilite ($\leqslant 3 \times 10^{-9}$) is consistent with the value of ^{60}Fe/^{56}Fe $\leqslant 4 \times 10^{-9}$ obtained by Chen et al. (2009) for troilite inclusions from other iron meteorites. Our estimate of the solar system initial ^{60}Fe/^{56}Fe further is lower than, but consistent with results from Regelous et al. (2008), who suggested an ^{60}Fe/^{56}Fe ratio $\leqslant 3 \times 10^{-7}$ for chondrites and $\leqslant 2.7 \times 10^{-7}$ for chondrules from Chainpur. Two ^{60}Fe–^{60}Ni isochrons for angrites suggest similar low solar system initial ^{60}Fe/^{56}Fe (Spivak-Birndorf et al. 2011; Quitté et al. 2010): an internal isochron for the D'Orbigny angrite (Spivak-Birndorf et al. 2011) and an external isochron for the three angrites NWA2999, SAH99555, and D'Orbigny (Quitté et al. 2010) both yield initial ^{60}Fe/^{56}Fe of $\approx 1 \times 10^{-8}$. These results also agree with a recent external isochron for eucrites, which gives an initial ^{60}Fe/^{56}Fe $\approx (4.90 \pm 2.70) \times 10^{-9}$ (Tang & Dauphas 2011). All these values of the initial ^{60}Fe/^{56}Fe ratio fall within the reported error bars of our results for Muonionalusta.

Although redistribution of Ni between metal and troilite during shocks and other secondary thermal events could have obliterated original ^{60}Ni excesses where previously present, the size of the inclusions analyzed here is large enough (>1 cm) to make diffusion inefficient. In addition, if shock had redistributed Ni over distances of several centimeters, Pb would certainly have been redistributed as well and the Pb–Pb age reset, which is not the case (Blichert-Toft et al. 2010a). Finally, Buchwald (1975) indicates that if shock melting happened in Muonionalusta, it was of very short duration.

^{60}Fe may be produced either from a single supernova explosion with injection of ^{60}Fe into the protoplanetary disk (Ouellette et al. 2007) or from the molecular cloud core progenitor (Cameron et al. 1995) of our solar system. AGB stars can produce ^{60}Fe with ^{60}Fe/^{56}Fe ratios up to 10^{-7}. However, the probability of having both a passing AGB star and a molecular cloud is very low (Kastner & Myers 1994) and AGB stars are usually not considered as viable sources of ^{60}Fe. Gounelle & Meibom (2008) surmised that the probability of direct injection of ^{60}Fe into the molecular cloud by a nearby star is very low ($\leqslant$1%). As an alternative to the explosion of a nearby supernova, Gounelle et al. (2009) suggested that ^{60}Fe was inherited from several supernovae belonging to previous episode(s) of star formation. This model requires a low initial ^{60}Fe/^{56}Fe ratio of the solar system (they used ^{60}Fe/^{56}Fe $= 3 \times 10^{-7}$). Our inferred ratio is even lower than that and, hence, weighs in favor of this model. The upper limit of the solar system initial ^{60}Fe/^{56}Fe ratio determined here does not, therefore, require the explosion of a nearby supernova to explain the origin of the solar system ^{60}Fe.

The absence of resolvable ^{61}Ni anomalies in the very old (or early) troilite inclusions of Muonionalusta as well as in troilite originating from the deep interior of the Nantan parent body, to where cosmic rays did not penetrate, is consistent with the lack of significant Ni isotope anomalies in IVA iron meteorites as a whole (Quitté et al. 2006; Cook et al. 2008). Likewise, our data on Mundrabilla troilite corroborate the deficiencies in ^{60}Ni and ^{61}Ni reported by Cook et al. (2008) for the same meteorite and confirm that non-magmatic iron meteorites (IAB-IIICD) currently are the samples with the largest Ni isotope anomalies.

Although the ^{61}Ni and ^{60}Ni anomalies reported so far in troilite inclusions from iron meteorites may be due to either cosmogenic (Quitté et al. 2006; Cook et al. 2008), nucleosynthetic, or spallation effects, we show in the following that these anomalies are unlikely to be due to cosmogenic effects. A number of different nuclear reactions induced by galactic cosmic rays potentially can change the ^{60}Ni/^{58}Ni ratios in iron meteorites but we expect the main reactions to be

1. production of ^{60}Co, primarily by neutron capture, and subsequent decay;
2. destruction (burn-out) of ^{58}Ni; and
3. low-energy spallation reactions on Cu producing ^{60}Ni.

The rates of each process can be estimated either from measurements of radionuclide activities or by modeling calculations.

Production of ^{60}Co by neutron capture. In iron meteorites, the radionuclide ^{60}Co is produced primarily, though not exclusively, by neutron capture on ^{59}Co. The ^{60}Co subsequently decays to ^{60}Ni:

$$^{59}\mathrm{Co}^{\star} + n \rightarrow {}^{60}\mathrm{Co}\,(t_{1/2} = 1925\,\mathrm{days}) \rightarrow {}^{60}\mathrm{Ni}. \quad (1)$$

The asterisk denotes production by neutrons. We assume that spallogenic production of ^{60}Fe, typically 1–2 dpm kg^{-1} (Knie et al. 1999), can be neglected in comparison with the production of ^{60}Co by thermal neutrons. Since essentially all ^{60}Co decays before the meteoroid ever gets to Earth, the production rate $P(^{60}\mathrm{Ni}^{\star})$ of ^{60}Ni by thermal neutron capture is virtually identical to the production rate of ^{60}Co by thermal neutron capture, $P(^{60}\mathrm{Co})$. In addition, because of its short half-life, ^{60}Co cannot be measured in iron meteorites with terrestrial ages of more than about 25 years, meaning that measurements are restricted to falls, which are rare.

Assuming constant irradiation conditions, the time-integrated production of ^{60}Ni* is

$$^{60}\mathrm{Ni}^{\star} = P(^{60}\mathrm{Co}) \times N(^{59}\mathrm{Co}) \times T_{\mathrm{exp}}, \quad (2)$$

where $N(^{59}\mathrm{Co})$ is the concentration of ^{59}Co and T_{exp} is the exposure age. In this formulation, $P(^{60}\mathrm{Co})$ has units of atom

^{60}Co (atom ^{59}Co)$^{-1}$ (time)$^{-1}$. The quantity of interest is the fractional change in the ^{60}Ni/^{58}Ni ratio. Assuming the initial abundances are terrestrial we have

$$\Delta(^{60}\mathrm{Ni}/^{58}\mathrm{Ni}) = \frac{P(^{60}\mathrm{Co}) \times N(^{59}\mathrm{Co}) \times T_{\mathrm{exp}}}{N(^{58}\mathrm{Ni})}. \quad (3)$$

Although rare, ^{60}Co activity measurements exist for a few meteorites. Measurements of activity, [A], effectively give $P(^{60}\mathrm{Co})$ $N(^{59}\mathrm{Co})$ but in units of (atom ^{60}Co) minute^{-1} (kg meteorite)$^{-1}$. To balance the units in Equation (3) we take T_{exp} in minutes and $N(^{58}\mathrm{Ni})$ in atom/(kg meteorite).

It should be understood that dpm ^{60}Co is equivalent to a number of decaying atoms ^{60}Ni* per minute.

Converting to standard measurement units, we have

$$\Delta(^{60}\mathrm{Ni}/^{58}\mathrm{Ni}) = \frac{A[\mathrm{dpm}/(\mathrm{kg\ meteorite})] \times T_{\mathrm{exp}}[\mathrm{Ma}]}{\mathrm{Ni\ [weight\ fraction]}} \times 7.530 \times 10^{-14}. \quad (4)$$

We take $10 \leqslant T_{\mathrm{exp}} \leqslant 1000$ (Ma) (Herzog 2003). The mass fractions of Ni and Co are 0.1 and 0.005, respectively (Malvin et al. 1984). Only a few ^{60}Co activities (production rates) have been measured on iron meteorites. Tobailem & Nordmann (1965) report a production rate of ^{60}Co of 14 ± 7 dpm kg^{-1} in the IAB Bogou; Honda & Arnold (1964) report a production rate of 95 ± 10 dpm kg^{-1} in Sikhote Alin (IIAB) and 17 ± 2 in Yardymly (IAB); and Song et al. (1976) report a production rate of 6.8 ± 2 dpm kg^{-1} in Ningbo (IVA). Using Equation (4), we calculated the ^{60}Ni production for the four iron meteorites Bogou, Sikhote Alin, Ningbo, and Yardymly to be less than 0.01 ppm (see Table 2). In addition, we computed an extreme case with low Ni and high Co concentrations and a maximum exposure age of 1.5 Ga. For the production rates, we used the maximum production rate of ^{60}Ni due to the decay of ^{60}Co reported by Kollar et al. (2006) for H chondrites of 100,000 atom ^{60}Co minute^{-1} [kg Co]$^{-1}$ and we get an effect of 1.5 ppm.

An independent way to calculate the production of ^{60}Ni by neutron capture on ^{59}Co is to model the ^{60}Co production rate in iron meteorites following the relationship:

$$^{60}\mathrm{Ni}^{\star} = \Phi \times N(^{59}\mathrm{Co}) \times \sigma(^{59}\mathrm{Co}), \quad (5)$$

where Φ is the total fluence of thermal neutrons, $N(^{59}\mathrm{Co})$ is the concentration of ^{59}Co, and σ is the neutron cross section.

Fluences are in the neighborhood of 1×10^{16} neutrons cm^{-2} (Shankar et al. 2011). For the cross section, we use the thermal and resonance integrals from Mughabghab et al. (1981), which are available online (http://ie.lbl.gov/ngdata/sig.txt) and are $\sigma = 1.11 \times 10^{-22}$ cm^2. With typical Co abundances in iron meteorites of 0.5 wt% (Malvin et al. 1984) and Ni abundances of 10 wt%, we get

$$^{60}\mathrm{Ni}^{\star}(\mathrm{atom}/[\mathrm{g\ meteorite}])/^{60}\mathrm{Ni}(\mathrm{atom}\,^{60}\mathrm{Ni}/[\mathrm{g\ meteorite}]) = 2.1 \times 10^{-7} = 0.2\ \mathrm{ppm}. \quad (6)$$

Relative to ^{58}Ni, we take the 0.2 ppm above and multiply by the abundance ratio $0.26/0.68 = 0.38$, to get 0.08 ppm, a value that compares with the measurements in iron meteorites (Table 2).

Nuclear reactions deplete ^{58}Ni and neutron capture, $^{58}\mathrm{N(n,\gamma)}^{59}\mathrm{Ni}$, is one important pathway:

$$^{58}\mathrm{Ni} + n \rightarrow\ ^{59}\mathrm{Ni}. \quad (7)$$

As it happens, ^{59}Ni is radioactive ($t_{1/2} = 760$ ka) and has been measured in some meteorites. These measurements, when taken together with cosmic-ray exposure ages, allow an estimate of ^{58}Ni loss assuming that irradiation conditions were constant and that corrections for terrestrial decay can be made. To a lesser degree, reactions such as $^{58}\mathrm{Ni}(n,\alpha)$ and $^{58}\mathrm{Ni}(p,\mathrm{x})$ may also play a role in destroying ^{58}Ni, but cross sections are probably lower by a factor of 10 or more than for the capture reaction. In the approximation that $^{58}\mathrm{Ni}(n,\alpha)\,^{59}\mathrm{Ni}$ is the most important reaction to consider, we have

$$\Delta^{58}\mathrm{Ni}[\mathrm{atom}/(\mathrm{kg\ meteorite})] = -A(^{59}\mathrm{Ni}\ [\mathrm{atom\ minute}^{-1} \times (\mathrm{kg\ meteorite})^{-1}] \times T_{\mathrm{exp}}(\mathrm{minutes}). \quad (8)$$

The fractional decrease in ^{58}Ni concentration is

$$\Delta(^{58}\mathrm{Ni}/^{58}\mathrm{Ni}) = \frac{-A[\mathrm{dpm}/(\mathrm{kg})] \times T_{\mathrm{exp}}[\mathrm{Ma}]}{W_{\mathrm{Ni}}[(\mathrm{kg\ Ni})/(\mathrm{kg\ meteorite})]} \times 7.530 \times 10^{-14}, \quad (9)$$

where W_{Ni} is the mass/weight fraction of Ni.

Typical ^{59}Ni activities (Honda & Arnold 1964) range from 50 to 100 dpm (kg meteorite)$^{-1}$. Thus, the shifts in ^{58}Ni are expected to be comparable to the shifts calculated above for ^{60}Ni but of opposite sign.

We can estimate a maximum combined effect of ^{60}Co production and ^{58}Ni destruction. The change in the ^{60}Ni/^{58}Ni ratio will be

$$^{60}\mathrm{Ni} = (^{60}\mathrm{Ni})_0 + P(^{60}\mathrm{Co})W_{\mathrm{Co}} \times T_{\mathrm{exp}}$$
$$^{58}\mathrm{Ni} = (^{58}\mathrm{Ni})_0 - P(^{59}\mathrm{Ni})W_{\mathrm{Ni}} \times T_{\mathrm{exp}}, \quad (10)$$

where W is a weight fraction in (kg element)/(kg meteorite), ^{x}Ni is a concentration in atom/(kg meteorite), and P is given in dpm/[kg element]. Hence we get

$$\epsilon\left(\frac{^{60}\mathrm{Ni}}{^{58}\mathrm{Ni}}\right) \approx \frac{P(^{60}\mathrm{Co}\ W_{\mathrm{Co}} \times T_{\mathrm{exp}}}{[\mathrm{Ni}] \times \mathrm{Abu}^{60}\mathrm{Ni}} + \frac{P(^{59}\mathrm{Ni})W_{\mathrm{Ni}} \times T_{\mathrm{exp}}}{[\mathrm{Ni}] \times \mathrm{Abu}^{58}\mathrm{Ni}}. \quad (11)$$

For this calculation, we assumed the maximum production rates of Kollar et al. (2006). For [Co] = 0.01, [Ni] = 0.10, and $T_{\mathrm{exp}} = 1000$ Ma, we get: $\epsilon\left(\frac{^{60}\mathrm{Ni}}{^{58}\mathrm{Ni}}\right) = 0.59$ ppm.

Low-energy spallation reactions on Cu producing ^{60}Ni. The importance of Cu is likely to be limited to selected phases in irons, possibly sulfides. The nuclear reactions of interest are $^{63}\mathrm{Cu(p,\alpha)}^{60}\mathrm{Ni}$ and $^{63}\mathrm{Cu(n,\alpha)}^{60}\mathrm{Co} \rightarrow\ ^{60}\mathrm{Ni}$. Cook et al. (2008) approximated these effects based on cross sections measured for reactions of protons with copper at a single energy. Here, we take as our model for these reactions the reaction $^{56}\mathrm{Fe(p\,\alpha)}^{53}\mathrm{Mn}$, ≈400 dpm/[kg Fe] with the reservation that the neutron-produced reactions are likely to be larger by a factor of two.

$$\Delta(^{60}\mathrm{Ni}/^{58}\mathrm{Ni}) = \frac{A[\mathrm{dpm}/(\mathrm{kg\ Fe})] \times W_{\mathrm{Cu}}[\mathrm{kg\ Cu}]/[\mathrm{kg\ meteorite}] \times T_{\mathrm{exp}}[\mathrm{Ma}]}{W_{\mathrm{Ni}}(\mathrm{kg\ Ni}/[\mathrm{kg\ meteorite}])} \times 7.530 \times 10^{-14}. \quad (12)$$

We adopt the following values:

$$A(^{53}\mathrm{Mn}) = 400\,\mathrm{dpm}/[\mathrm{kg\ Fe}] \approx 400\,\mathrm{dpm}/[\mathrm{kg\ meteorite}]$$
$$W_{\mathrm{Cu}} = 0.01\,\mathrm{kg\ Cu}/[\mathrm{kg\ meteorite}]$$
$$T_{\mathrm{exp}} = 500\,\mathrm{Ma}$$
$$W_{\mathrm{Ni}} = 0.05\,\mathrm{kg\ Ni}/[\mathrm{kg\ meteorite}],$$

Table 2
Isotopic Effect of Neutron Capture on ^{59}Co on the ^{60}Ni/^{58}Ni Ratio

Sample Name	Ni Weight Fraction	Co Weight Fraction	Reference	T_{exp} Ma	Reference	^{60}Co (dpm kg^{-1})	Reference	$\Delta(^{60}$Ni/^{58}Ni) ppm[h]
Bogou (IAB)	0.0733	0.0047	a	666	b	14	c	0.0098
Sikhote Alin (IIAB)	0.0587	0.005	d	132	b	95	e	0.016
Yardymly (Aroos) (IAB)	0.075	0.00495	a	100	b	17	e	0.016
Ningbo (IVA)	0.082	0.0038	f	110	b	7	g	0.00071
Maximum[f]	0.05	0.01	...	1000	...	0.01×10^5	...	1.5

Notes.
[a] Wasson & Kalleymen (2002).
[b] Herzog (2003).
[c] Tobailem & Nordmann (1965).
[d] Wasson (1969).
[e] Honda & Arnold (1964).
[f] Malvin et al. (1984).
[g] Song et al. (1976).
[h] Values chosen at the extremes of the respective ranges so as to maximize $\Delta(^{60}$Ni/^{58}Ni).

which gives $\Delta(^{60}$Ni/^{58}Ni$) = 0.003$ ppm. As noted above, neutron reactions probably contribute a factor of 4–10 more ^{60}Ni, which may bring the total up to 0.03 ppm.

In summary, our calculations show that production of ^{60}Ni by spallation during the exposure history of iron meteorites is limited to excesses of up to 1.5 ppm and, hence, does not account for the change in the ^{60}Ni/^{58}Ni ratio observed for Mundrabilla, which is a depletion larger than 100 ppm. In line with Cook et al. (2008), we therefore conclude that this anomaly represents an incompletely homogenized mixture of nebular material with stellar nucleosynthetic products from a Type II supernova. High-precision Ni isotope data for two troilite inclusions from Muonionalusta show no resolvable deviation from the terrestrial standard. In particular, the radiogenic isotope ^{60}Ni shows no excesses at the level of the measurement precision of 10 ppm. From these new isotopic data and the corresponding Fe/Ni ratio, an initial ^{60}Fe/^{56}Fe ratio of less than 3.2×10^{-9} is derived. The Muonionalusta troilite has been precisely dated to be 4565.3 ± 0.1 Ma old and, thus, assuming that the ^{60}Fe/^{56}Fe ratio was homogenously distributed in the early solar system, our results suggest a solar system initial ^{60}Fe/^{56}Fe ratio of 5×10^{-9}, which is in agreement with recent solution work (Tang & Dauphas 2011; Chen et al. 2009) but lower than in situ measurements (Tachibana et al. 2006; Mostefaoui et al. 2005) by at least a factor of 50.

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