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Ni isotope systematics in chondrules from unequilibrated chondrites to constrain ^{60}Fe in the early Solar System

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Presence of ^{60}Fe in early Solar System was established more than two decades ago, but the Solar System initial ratio of $^{60}Fe/^{56}Fe$ is, still, not well constrained. Isotopic studies of bulk meteorites samples from differentiated meteorites, achondrites, and chondrules suggest a low Solar System initial $^{60}Fe/^{56}Fe$ value of ~2×10^{-8}, while the *in-situ* studies using secondary ion mass spectrometer suggest a much higher Solar System initial value of ~7×10^{-7}.

XII International Symposium on Nuclei in the Cosmos
August 5-12, 2012
Cairns, Australia

[1] Speaker

1. Introduction

Understanding and constraining the sequence of events in the first 10 Myrs history of the Solar System is crucial for deciphering the origin of the Solar System and its subsequent evolution. Short-lived, now-extinct, radionuclides (SLNs), with a half life in the range of 0.01-86 Myrs, provide relative chronology with a high temporal resolution that is not possible with an absolute chronometer, except for U-Pb in favourable cases. These SLNs provide relative chronology of early Solar events. The SLNs found in various components of meteorites can be products of 1) Stellar nucleosynthesis, 2) Energetic particle interactions with gas and dust present in proto-Solar molecular cloud, or 3) Galactic chemical evolution.

Since various processes and sources can produce different amount of SLNs, abundances of these nuclides provide additional constraints on the source of SLNs. While many SLNs, like ^{41}Ca, ^{26}Al, can be produced by both stellar nucleosynthesis and energetic particle interactions, there are a few nuclides like ^{10}Be, ^{7}Be, and ^{60}Fe which are produced effectively only uniquely by one of the processes. ^{10}Be, and ^{7}Be are produced by spallation reactions while ^{60}Fe is produced by a stellar source. Hence, nickel isotopic studies in early Solar System solids can provide essential constraints on the stellar source of SLNs.

The former presence of ^{60}Fe with an upper limit ^{60}Fe/^{56}Fe value of ~1×10^{6} in early Solar System was first suggested following study of nickel isotopes in Allende CAIs [1]. Confirmatory evidence of a correlated excesses in ^{60}Ni with Fe/Ni ratios suggestive of in-situ decay of ^{60}Fe was shown in the bulk samples of Chervony Kut Eucrite, although mineral separate of this meteorite showed perturbed isotopic records [2]. The initial efforts to find fossil records of ^{60}Fe were made using in-situ studies using secondary ion mass spectrometry in the earliest formed solids initially and in bulk meteorite samples using multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS) in recent years. The initial attempts in CAIs, and chondrules were not successful and only upper limits could be inferred. Subsequent efforts in Fe-rich phases: troilites, and magnetites in matrix yielded a ^{60}Fe/^{56}Fe value of ~1-9×10^{-7} [3,4]. Nickel being quite labile in metal, and sulphide phases and the lack of an independent estimate of time of formation of sulphides and magnetites did not allow an unequivocal estimation of Solar System initial (SSI) ^{60}Fe/^{56}Fe. Subsequently, study of Fe-Ni isotope systematics in silicates, which are more robust to thermal perturbation, yielded ^{60}Fe/^{56}Fe in the range of ~2×10^{-7} [5-8]. However, recently the results from some of the in-situ studies have been revised to show no excess owing to the error in the data reduction techniques involving low counts [9]. Efforts have also been made to detect excesses in bulk samples using high precision MC-ICPMS [10-18]. Currently, there seems to be discordance between the results obtained from SIMS which suggest an order of magnitude higher value compared to the results from MC-ICPMS.
^{60}Fe-^{60}Ni isotope systematics in silicates.

Calcium-, aluminium rich inclusions (CAIs) are amongst the first solids in the Solar System. Hence, principally they are the prime objects of interest to infer the SSI abundance of ^{60}Fe in the early Solar System, but the following characteristics of CAIs reduce their utility for unambiguous inferences: 1) The refractory minerals constituting the CAIs do not incorporate moderately volatile Fe resulting in a low Fe/Ni ratios close to the Solar value making detection of excess in ^{60}Ni challenging using presently available analytical techniques. 2) CAIs host nucleosynthetic anomalies in n-rich isotopes of elements from Mn-Cu. Hence, it is difficult to estimate the contribution of each component and delineate the excess in ^{60}Ni resulting from in-situ decay of ^{60}Fe. 3) Petrography of CAIs suggest that their formation and subsequent evolution has been through at least a few high temperature processes involving evaporation, condensation, open/close system interaction with ambient gas exchange resulting in convoluted isotopic records.

Hence, the other early formed Solar System solids –chondrules that are devoid of the above lacunae are more suitable to search for extinct ^{60}Fe. None-the-less, unambiguous detection is analytically challenging owing to rarity of high ^{56}Fe/^{62}Ni ratios in silicate phases in chondrules. A study of ^{60}Fe-^{60}Ni, and ^{26}Al-^{26}Mg isotope systematics within chondrules can give the abundance of ^{60}Fe (^{60}Fe/^{56}Fe) at the time of formation of chondrules and an independent estimate of time of formation of chondrules from ^{26}Al-^{26}Mg isotope systematics; thus allowing an estimate of the SSI ^{60}Fe/^{56}Fe. However, while porphyritic chondrules have large glassy phases with high Al/Mg suitable for Al-Mg isotope systematics, they have paucity of high Fe/Ni ratios in silicate phases, on the other hand non-porphyritic chondrules host high Fe/Ni phases, but do not have large Al-rich phase making such a study challenging to perform. Hence, SSI ^{60}Fe has been inferred in most studies by assuming an approximate time of formation of the analysed objects. Here we discuss the progress made and unresolved issues concerning ^{60}Fe records in meteorites. The results from various studies, where the presence of ^{60}Fe has been reported, are shown in Table 1 and Fig. 1.

2. ^{60}Fe-^{60}Ni isotope systematics using SIMS: A typical analytical protocol utilises an O^- primary beam of ~ 15-25µm spot size carrying ~ 10-20 nA, to produce secondary positively charged ions of ^{57}Fe, ^{60}Ni, ^{61}Ni, and ^{62}Ni. A mass resolution of ~ 4000 resolves all the major molecular interferences, except for hydrides. Ion intensities of 56.5, ^{57}Fe, ^{60}Ni, ^{61}Ni, and ^{62}Ni are measured in peak jumping pulse counting mode using electron multipliers in mono/multi-collection mode for suitable time period to obtain statistically significant data. The estimation of dynamic background at 56.5 is essential to make an appropriate correction in view of the low counts in high Fe/Ni phases. In cases with low count rates, typically for high Fe/Ni regions, sum of counts is taken to estimate the ratios. All the reported excesses in sulphides, and silicates from the in-situ SIMS analysis, except for 3 chondrules, by the Hawaii group have been revised downward to show no excess owing to a statistical bias in the protocol of data reduction [9].

In another study, a set of carefully chosen chondrules from unequilibrated ordinary chondrites of Semarkona (LL 3.0), LEW 86134 (L 3.0), and Y 791324 (L 3.05) shows well resolved excesses in ^{60}Ni* in about ten chondrules at 2 sigma level [6-8,19]. Seven of these chondrules also show well resolved excesses in ^{26}Mg* as shown in Fig. 1. Mishra and Chaussidon 2012 reported well resolved excesses in ^{60}Ni* in two additional chondrules from Semarkona and also from the only type II chondrule present in a thin section of Efremovka [20,21]. These 3 chondrules have ^{60}Fe/^{56}Fe in the range of $2\text{-}8\times10^{-7}$ consistent with the earlier study. However, ^{26}Al/^{27}Al values for these chondrules are not presently known. In another two chondrules from QUE 97008 (L 3.05), well resolved excesses at 2 sigma level in the range of $2\text{-}7\times10^{-7}$ have been reported from a NanoSims study of silicate phases in chondrules [22]. All the data from in-situ studies of silicate phases in chondrules and sulphides showing resolved excesses are shown in Fig. 1. The abundances of ^{60}Fe in seven chondrules that also have time of formation inferred

Table 1. Initial $^{60}Fe/^{56}Fe$ from various components of meteorites in different studies.

Year	Authors & journal	Meteorite(s)	Mineral(s)	Result $^{60}Fe/^{56}Fe$	Analytical Conditions
2012	Mishra & Chaussidon Metsoc., NIC	Efremovka, Semarkona	Olivine, Pyroxene	$(7.8\text{-}2.2)\times10^{-7}$	SIMS 1270
2012	Marhas & Mishra Metsoc.	Que 97008	Olivine, Pyroxene	$(7.7\text{-}3.4)\times10^{-7*}$ $(10\text{-}6)\times10^{-7}$	NanoSIMS
2012 2011	Telus et al. LPSC, MetSoc.,	Semarkona, Bishunpur, Krymka,	Olivine, Pyroxene, Sulphide	$(2.4\text{-}0.6)\times10^{-7}$	SIMS 1280
2012	Tang & Dauphas LPSC	Achondrites Semarkona, NWA5717, Chainpur, Gujba (CB)	WR, Min. Separate	$(1.38\pm0.4)\times10^{-8}$ $(3.2\pm0.53)\times10^{-9}$ $(1.5\pm0.97)\times10^{-8}$ **$(2.3\pm1.6)\times10^{-8*}$**	MC-ICPMS
2012	Spivak et al. MetSoc	Semarkona, Que 97008, Chainpur	WR	$(.74\pm0.3)\times10^{-8}$ $(1.3\pm0.6)\times10^{-8}$	MC-ICPMS
2011	Quitte et al., GCA 75	Juvinas, Bouvante	WR, Min. Sep	$(0.4\text{-}\ 5)\times10^{-9}$	MC-ICPMS
2010	Mishra et al. ApJ 714	Semarkona, Bishunpur, LEW 86134	Pyroxene, Olivine	**$(6.3\pm2.0)\times10^{-7}$**	SIMS 4f
2010	Quitte et al., APJ 720	NWA2999, D'Orb, Sah 99555	WR, Min. Sep	$(1\text{-}\ 7)\times10^{-9}$	MC-ICPMS
2008	Dauphas et al. ApJ 686	IIAB, IIIAB, PMG,Bishunpur	Metals, Bulk	$< 6\times10^{-7}$	MC-ICPMS
2007	Quitte et al., APJ 655	Allende, Efremovka	CAIs Bulk	$(4.2\text{-}\ 4.7)\times10^{-7}$	ICPMS
2005	Mostefaoui ApJ 625	Semarkona	Troilite, Magnetite	$(9.2\pm2.4)\times10^{-7}$ $(1.1\pm0.7)\times10^{-7}$	NanoSIMS
1993	Shukolyukov & Lugmair Science, 259	Chervony Kut	Troilite, Olivine	$(3.9\pm0.6)\times10^{-9}$	TIMS
1988	Birck & Lugmair EPSL, 90	Allende	CAIs Bulk	$<1.6\times10^{-6}$	TIMS

from Al-Mg isotope systematics gives a weighted average SSI $^{60}Fe/^{56}Fe$ value of $(7.0\pm1.2)\times10^{-}$

[7] [19]. Recentdevelopments in analytical techniques have also led to search for fossil records of ^{60}Fe in bulk
samples and mineral separates using MC-ICPMS by various groups. The bulk rock isochrons of cumulate and non cumulate eucrites yield $^{60}Fe/^{56}Fe$ ~$(3.0\pm1.5)\times10^{-9}$. The angrites also yield a similar value of ~$(3.5\pm1.5)\times10^{-9}$. Considering ^{53}Mn-^{53}Cr dating of these objects a SSI $^{60}Fe/^{56}Fe$ ~ $(1.5\pm1.0)\times10^{-8}$ has been inferred [16-18]. Tang and Dauphas 2012, and Spivak et al. 2012 have also reported the bulk isochrons of chondrules from Semarkona, Chainpur, QUE 97008, NWA 5717 and suggest a $^{60}Fe/^{56}Fe$ ratio in the range of ~ $(0.74 - 1.52)\times10^{-8}$.

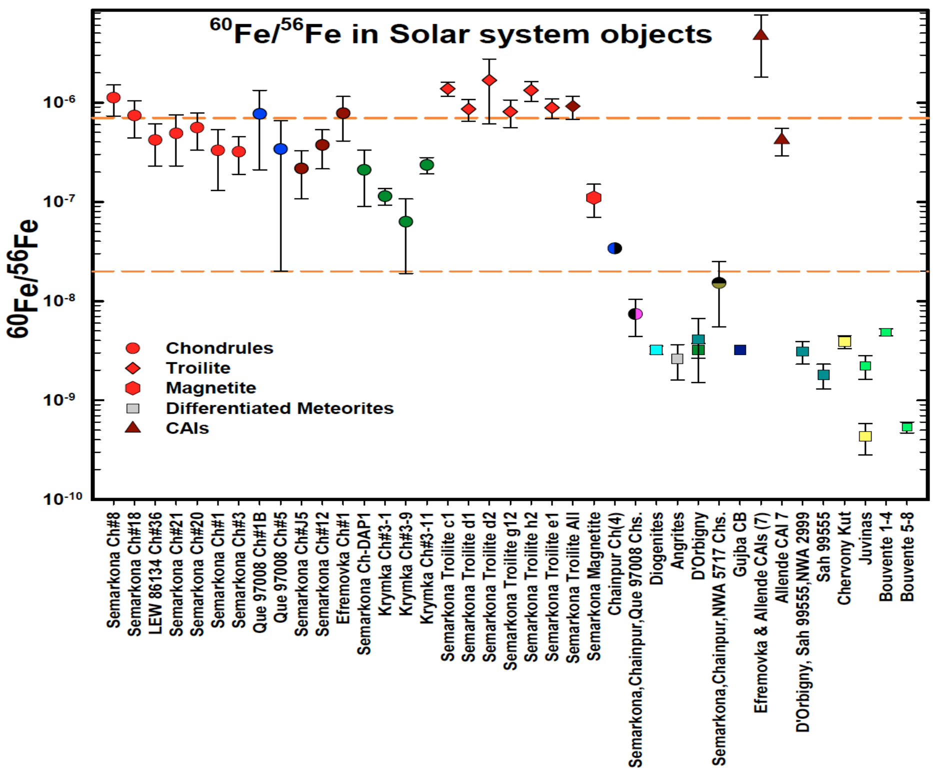

Fig. 1. Various Solar System objects showing a range of $^{60}Fe/^{56}Fe$ values. Data from chondrules (circle), CAIs (triangle), troilites (rhombus), and differentiated meteorites (squares). The thick and thin dashed lines represent the suggested Solar System initial $^{60}Fe/^{56}Fe$ from in-situ studies and bulk studies.

3. Discussion: The present data obtained from in-situ studies using SIMS and bulk isochrons from MC-ICPMS seem discordant at their face values. The inferred SSI $^{60}Fe/^{56}Fe$ values from these studies differ by a factor of ~ 40. The bulk data obtained using MC-ICPMS are precise indeed and their utilisation of samples that have been significantly fractionated in Fe/Ni due to differentiation should yield a consistent value from other Solar System objects. The results from differentiated objects seem to be consistent amongst themselves. However, it has been observed that the mineral isochrons and in some case even bulk isochrons are disturbed and possibly represent a mixing line [13,14]. Most of the bulk data from chondrules have Fe/Ni close to the Solar value and with precision obtained from the current analytical techniques it should be difficult to discern radiogenic excesses. Incidently, the bulk isochrons of chondrules have a much lower intercept inconsistent with the expectation of low SSI $^{60}Fe/^{56}Fe$. The concert study of Al-Mg, and Fe-Ni within the same chondrule of about a dozen chondrules from least altered meteorites showing a correlation between ^{26}Al and ^{60}Fe despite larger errors give greater credence to a higher Solar System ^{60}Fe/^{56}Fe. In addition, MC-ICPMS study of seven bulk CAIs, though having low Fe/Ni values, give a higher SSI ^{60}Fe/^{56}Fe ~ $(4.7\pm2.9)\times10^{-6}$. In particular, an Allende CAI (#7) which does not show a signature of nucleosynthetic anomaly gives a SSI ~ $(4.2\pm1.3)\times10^{-7}$ which is consistent with those inferred from the chondrules. However, the results obtained from in-situ and bulk need to be reconciled, possibly by analysing the same sample using both techniques.

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