CLEAR EVIDENCE FOR ^{60}FE IN SILICATE FROM A SEMARKONA CHONDRULE.

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Introduction: ^{60}Fe ($t_{1/2}$ = 1.5 Ma) is key to understanding the sources of short-lived radionuclides in the early solar system because it is the only one among those known from meteoritic material that is produced only in stars [1]. Within the last year, it has become clear that ^{60}Fe was present in sulfides from primitive ordinary and enstatite chondrites in amounts sufficient to require a recent stellar input [2-5]. The sulfide data indicate an initial $^{60}Fe/^{56}Fe$ ratio for the early solar system of between ~3×10^{-7} and ~1.6×10^{-6} [2–4]. However, iron (and nickel?) in sulfides is easily mobilized by very mild heating [e.g., 6], so there is considerable uncertainty over the true initial ratio. To resolve this uncertainty, we have begun a search for evidence of ^{60}Fe in silicates from primitive chondrites. In olivine from type 3.0-3.1 ordinary chondrites, diffusive exchange of iron and magnesium has not occurred to any significant degree, and diffusive exchange in pyroxene is slower [7]. However, the relatively small elemental fractionation of iron from nickel in silicates, coupled with the fact that the daughter nuclide, ^{60}Ni, makes up ~26 % of normal nickel, make detection of excesses of radiogenic ^{60}Ni very difficult. Fortunately, we have found a fine-grained radiating-pyroxene chondrule in Semarkona (LL3.0) with a very high Fe/Ni ratio that gives clear evidence of ^{60}Fe.

Experimental: Several chondrules with iron-rich olivine or other high-iron phases were selected and documented in the SEM. Isotopic measurements were made with the ASU Cameca ims 6f ion microprobe. A focused ~3.3 nAmp primary O^- beam was used to generate positive iron and nickel ions. The secondary mass spectrometer was operated at 8.8 KeV with a 60 eV energy window and a mass resolving power of ~5000, sufficient to resolve all interferences except for hydrides on $^{57}Fe^+$, $^{61}Ni^+$, and $^{62}Ni^+$. Hydrides were suppressed by baking the system prior to analysis and with a liquid nitrogen trap. High-resolution mass scans showed that the hydride contribution at $^{61}Ni^+$ was <1‰ on all samples. Data are corrected for electron multiplier background (0.014 cps) and dead-time (24 nsecs). San Carlos olivine was the nickel isotope standard, and San Carlos olivine and Famous 519-4 mid-ocean ridge basalt were used to estimate the Fe/Ni sensitivity factor. The data were reduced in several ways to assure that results are not artifacts of the data reduction. 1) Instrumental mass fractionation was accounted for internally for each point using the measured $^{62}Ni/^{61}Ni$ ratio. Instrumental fractionation was also corrected for externally using 2) the average $^{60}Ni/^{61}Ni$ of the standards, and 3) the average $^{60}Ni/^{62}Ni$ of the standards. Nickel and iron isotope ratios are from [8, 9]. Each spot was measured for two hours. Because nickel count rates are so low, many measurements were required to achieve statistically significant results, so plotted data are the weighted means of several analyses with similar Fe/Ni ratios.

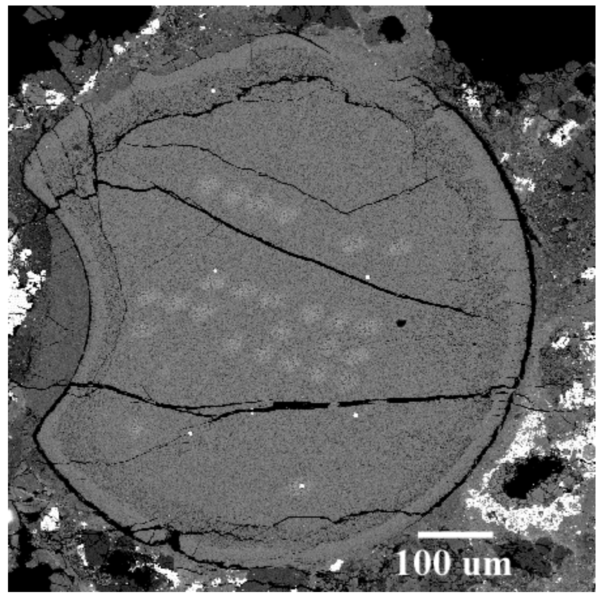

Fig. 1: BSE image of fine-grained radiating pyroxene chondrule from Semarkona showing evidence for ^{60}Fe. Light-colored spots are ion-probe pits.

Results: Isotopic data were collected on three Semarkona chondrules. Two are porphyritic olivine chondrules with large clean olivine crystals and one is a large, fine-grained, radiating pyroxene chondrule (Fig. 1). The two porphyritic chondrules did not have high enough Fe/Ni ratios (Fe/Ni ≈ 2400, 1100) to permit detection of radiogenic ^{60}Ni. The upper limits on the initial $^{60}Fe/^{56}Fe$ ratio in these chondrules are $<3\times10^{-6}$ and $<6\times10^{-6}$. These ratio are considerably higher than the upper limit of ~3.4×10^{-7} established by [10] for a type II, olivine-bearing, Semarkona chondrule with Fe/Ni ≈ 2400. In contrast, the fine-grained radiating pyroxene chondrule has Fe/Ni ratios up to >20,000 and does show evidence of radiogenic ^{60}Ni that correlates with Fe/Ni ratio (Fig. 2). When the data are corrected internally for instrumental mass fractionation using the measured $^{62}Ni/^{61}Ni$ ratios, the inferred initial $^{60}Fe/^{56}Fe$ ratio is $(3.1\pm1.5)\times10^{-7}$. When corrected externally using either the $^{60}Ni/^{61}Ni$ or $^{60}Ni/^{62}Ni$ ratios of the standards, the inferred initial $^{60}Fe/^{56}Fe$ ratios are $(1.8\pm0.9)\times10^{-7}$ and $(2.2\pm1.1)\times10^{-7}$, respectively. Thus, the identification of excess ^{60}Ni does not depend on the correction method, but the data are not precise enough to constrain the initial $^{60}Fe/^{56}Fe$ ratio to better than $\sim(1–4)\times10^{-7}$, with a preferred value of $\sim2.4\times10^{-7}$. This value is compatible with, but perhaps slightly higher than, the value inferred by [2,3] for troilites from Krymka and Bishunpur, but is significantly lower than the value obtained from Semarkona troilite by [5].

Discussion: The previously collected sulfide data on primitive chondrites has proved difficult to interpret. Apparent isochrons for Krymka sulfides give initial $^{60}Fe/^{56}Fe$ ratios of $(1.6–1.8)\times10^{-7}$ [2,3], while a Semarkona troilite gave $\sim8\times10^{-7}$ [4]. Sulfides from enstatite chondrites give apparent initial ratios ranging from a few $\times10^{-7}$, consistent with the UOC sulfide data, to $\sim10^{-5}$ [5]. This latter ratio is too high to be credible. The highest ratios are most likely the result of iron loss after the decay of ^{60}Fe, which steepens the isochrons. It is clear both from previously collected data [5] and from experimental studies [6] that iron, at least, is very mobile in sulfides at low temperature [cf, 3]. Thus it is impossible to be sure that any of the apparent isochrons obtained on sulfides is undisturbed. Silicates are less subject to diffusive re-equilibration than sulfides, and in type 3.0-3.1 ordinary chondrites the silicates may have survived unaltered since their formation [7]. This is why we are attempting the much more difficult experiment of measuring silicates in chondrules. Other phases, such as magnetite, may also be suitable.

If our Semarkona chondrule is undisturbed and if we have obtained an accurate estimate of its initial ratio, then we can attempt to infer $(^{60}Fe/^{56}Fe)_0$ for the solar system. The $^{60}Fe/^{26}Al$ ratio of the early solar system constrains the potential stellar source(s) for the short-lived radionuclides [e.g., 11]. A hard lower limit comes from the measured ratio, which could be as low as $\sim1\times10^{-7}$; this assumes that the chondrule formed at time zero. However ^{26}Al systematics indicate that Semarkona chondrules formed 1-2 Ma after the CAIs [10]. Assuming this time delay and taking our upper limit on the initial ratio for the Semarkona chondrule ($\sim4.6\times10^{-7}$), we calculate an upper limit on $(^{60}Fe/^{56}Fe)_0$ of $\sim9\times10^{-7}$. However, our most probable value for $(^{60}Fe/^{56}Fe)_0$, based on an inferred initial $^{60}Fe/^{56}Fe$ ratio for our chondrule of $\sim2.5\times10^{-7}$, is $\sim5\times10^{-7}$, which is slightly higher than the $(2.8–4)\times10^{-7}$ inferred by [2, 3] from UOC troilite and considerably lower than the $\sim(1.6–2)\times10^{-6}$ that would be inferred from the Semarkona troilite data [4] using similar assumptions. We are continuing our search for chondrules suitable for ^{60}Fe measurements.

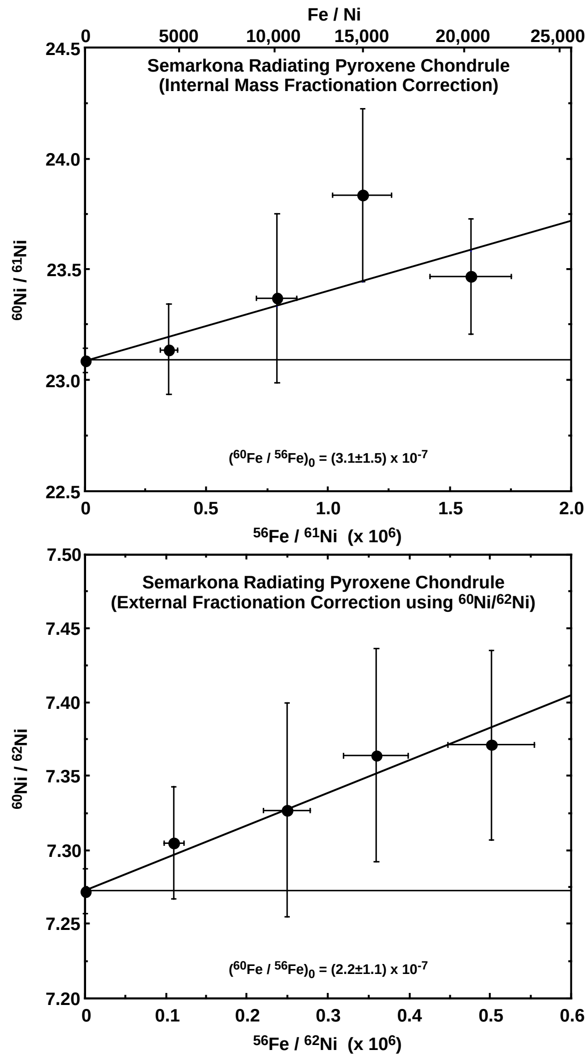

Fig. 2: Isochron diagram showing data for radiating pyroxene chondule in Fig. 1. Individual two-hour measurements have been combined into weighted means, which are plotted here.

References: [1] Lee T. et al. (1998) *Ap. J.* 506, 898-912. [2] Tachibana S. and Huss G. R. (2003) *LPS XXXIV*, Abstract #1737. [3] Tachibana S. and Huss G. R. (2003) *Ap. J. 588,* L41-L44. [4] Mostefauoi S. et al. (2003) *LSP XXXIV*, Abstract # 1585. [5] Guan Y., Huss G. R., Leshin L. A. and MacPherson G. J. (2003) *Meteorit. Planet. Sci. 38,* A138. [6] Condit R. H. et al. (1974) *Oxid. Metals 8*, 409-455. [7] DeHart J. M. et al. (1992) *GCA 56*, 3791-3807. [8] Birck J. L. and Lugmair G. W. (1988) *EPSL 90*, 131-143. [9] Völkening J. and Papanastassiou D. A. (1989) *Ap. J. 347* L43-L46. [10] Kita N. T. et al. (2000) *GCA 64*, 3913-3922. [11] Wasserburg G. J. et al., (1998) *Ap. J. 500*, L189-L192. Supported by NASA NAG5-11543.