

**INITIAL ABUNDANCE OF  $^{60}\text{Fe}$  IN UNEQUILIBRATED ORDINARY CHONDRITES.** M. Telus, G. R. Huss, K. Nagashima, R. C. Ogliore, HIGP, Univ. of Hawai'i at Mānoa, Honolulu, HI 96822 [telus@higp.hawaii.edu](mailto:telus@higp.hawaii.edu).

**Introduction:** The  $^{60}\text{Fe}$ - $^{60}\text{Ni}$  system ( $t_{1/2} = 2.6$  Myr) can potentially provide important constraints on early solar system chronology, the stellar source of short-lived radionuclides (SLRs), and the environment and conditions of the solar system's formation. An accurate and precise initial  $^{60}\text{Fe}/^{56}\text{Fe}$  ratio in the solar system,  $(^{60}\text{Fe}/^{56}\text{Fe})_{\text{ss}}$ , is necessary to address these topics. High values for the  $(^{60}\text{Fe}/^{56}\text{Fe})_{\text{ss}}$  of up to  $\sim 10^{-6}$  have been inferred based on ion microprobe measurements of sulfides and silicates from chondritic material [1-5]. The presence of  $^{60}\text{Fe}$  at this level in the early solar system has been used to support a supernova origin for short-lived radionuclides [6,7].

We have recently shown that much of the published ion probe Fe-Ni isotopic data were biased due to improper data reduction [4,8,9]. This bias correlates inversely with the number of the counts of the normalizing isotope. When counts in the denominator isotope are low (as in the case of our Ni measurements), the biased ratios produce correlations that look like isochrons [8]. We have now published recalculated SIMS data for various SLR systems, including most of the previously published Fe-Ni data [9]. After correcting the data, most of the originally reported high initial  $^{60}\text{Fe}/^{56}\text{Fe}$  ratios disappeared or were much lower than the original estimates. Initial  $^{60}\text{Fe}/^{56}\text{Fe}$  ratios at these lower levels significantly weaken the requirement for a supernova source of short-lived radionuclides.

Using our improved analytical technique and appropriate data reduction methods, we can confidently approach analyzing new samples. Here, we discuss our recent Fe-Ni measurements of chondrules from primitive ordinary chondrites and their implications.

**SIMS Technique:** We measured Fe and Ni isotopes in multicollection mode using the Cameca ims 1280 ion microprobe at the University of Hawai'i. We used a  $\sim 10$  nA  $^{16}\text{O}^-$  beam with a  $15\ \mu\text{m}$  raster for each measurement. Nickel isotopes,  $^{60}\text{Ni}^+$ ,  $^{61}\text{Ni}^+$ , and  $^{62}\text{Ni}^+$ , were measured simultaneously on electron multipliers (EMs) with a B-field jump to  $^{57}\text{Fe}^+$ , which was measured on a Faraday cup. Before each measurement, we presputtered each spot with a  $20\ \mu\text{m}$  raster for 300 s. Molecular interferences on the Ni isotopes (e.g.,  $^{44}\text{Ca}^{16}\text{O}$ ,  $^{45}\text{Sc}^{16}\text{O}$ , and  $^{46}\text{Ca}^{16}\text{O}$ ) were monitored at the end of each measurement. A mass resolving power (MRP) of  $\sim 4800$  mostly resolves these interferences, but we still had to make a small tail correction. We used a synthetic low-Ca pyroxene standard for standard-sample bracketing and for monitoring EM drift.

**Data Analysis:** Corrections were made for background, deadtime, and drift in the EMs. Counts from

the tails of interferences were corrected based on abundance sensitivity measurements on the standard ( $\sim 50$  ppm). Instrumental mass fractionation was corrected internally for each individual measurement. Isotope ratios were calculated from the total counts acquired for each isotope to avoid complications due to ratio bias [8]. The number of total counts for the normalizing isotope was monitored to assure that any residual bias is insignificant.

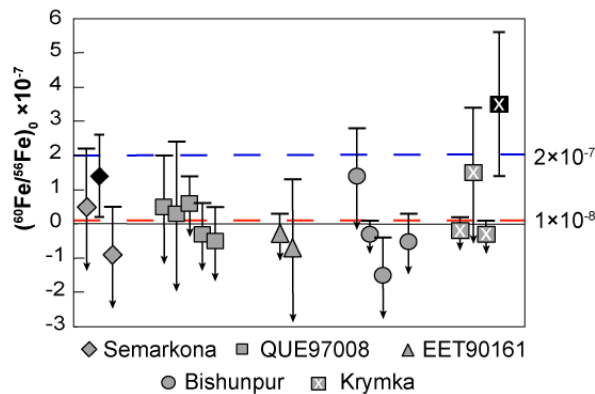
**Sample Selection:** The best samples for Fe-Ni measurements are those that have experienced little to no aqueous alteration or thermal metamorphism. We analyzed low-Ca pyroxene grains in 5 chondrules from 3 different ordinary chondrites, including two pyroxene-olivine porphyritic (POP) chondrules from QUE97008 (L3.05), a barred pyroxene chondrule and a POP chondrule from EET90161 (L3.05), and a barred pyroxene chondrule from Semarkona (LL3.00). Both QUE97008 and EET90161 were characterized as pristine chondrites of petrologic type 3.05 based on the Cr distribution in Fe-rich olivines [10]. Raman spectroscopic measurements of the matrix in these meteorites made using the protocol of [11] give results consistent with this classification. These samples are the best we have available, but thermal alteration of the Fe-Ni system could still be significant.

**Results:** Preliminary results for the chondrules we analyzed most recently are presented in Table 1. We did not find clear evidence for the former presence of  $^{60}\text{Fe}$  in these samples, so we report only upper limits ( $2\sigma$ , 2-sided) on the initial  $(^{60}\text{Fe}/^{56}\text{Fe})_0$  ratios.

**Table 1.** Preliminary results for recent measurements.

Sample	$^{56}\text{Fe}/^{62}\text{Ni} \times 10^5$	$(^{60}\text{Fe}/^{56}\text{Fe})_0 \times 10^{-7}$	$\chi_v^2$
QUE ch K	0.3-1.4	< 1.2	1.5
QUE ch I	0.1-1.6	< 2.4	0.9
EET r1	1.0-8.2	< 0.3	1.6
EET r4	0.3-1.5	< 1.3	1.0
SMK r1	0.4-1.0	< 2.4	1.2

The initial  $^{60}\text{Fe}/^{56}\text{Fe}$  ratios for the various chondrules we have measured during the past couple of years are plotted in Figure 1. Only two chondrules in Figure 1 have resolved initial  $^{60}\text{Fe}/^{56}\text{Fe}$  ratios (symbols in black). Both are radiating pyroxene chondrules. The regressions for these two chondrules show weak correlations, and have  $\chi_v^2$  of 11 and 3.3 for Semarkona and Krymka, respectively, indicating that the Fe-Ni system in these chondrules is disturbed.



**Fig. 1.** Inferred  $(^{60}\text{Fe}/^{56}\text{Fe})_0$  ratios for chondrules in type 3.0-3.2 UOCs. Symbols in black are for chondrules with resolved initial ratios. All uncertainties are  $2\sigma$ .

**Discussion:** ICPMS measurements of UOC chondrules give  $(^{60}\text{Fe}/^{56}\text{Fe})_0$  ratios of  $\sim 1 \times 10^{-8}$  [12-14]. Can our results be reconciled with these values? With the exception of the two chondrules that appear to give resolved initial ratios of  $(1-3) \times 10^{-7}$ , all of our data are consistent with the ICPMS values. However, many of them also permit initial ratios up to  $\sim 1 \times 10^{-7}$  and a few permit ratios of  $\sim 2 \times 10^{-7}$ .

To constrain the initial  $(^{60}\text{Fe}/^{56}\text{Fe})_{\text{ss}}$ , we have focused our efforts on measuring chondrules from the most-pristine ordinary chondrites. However, it is well known that even Semarkona, the most pristine of the UOCs, experienced some degree of thermal processing with a peak temperature of up to 200-250 °C [15,16]. Krymka (LL3.2) may have reached  $\sim 400$  °C [16]. Chromium begins to leave Fe-rich olivine in type 3.05 chondrites and has mostly left the olivine by type 3.1/3.2 [10]. Experimental data on Ni diffusion shows that Ni diffuses  $\sim 3 \times$  faster than Cr in olivine [17]. This implies that the Fe-Ni system in primitive UOC olivine was not closed, even in Semarkona. We focused our measurements on pyroxenes because they are relatively resistant to metamorphism [18]. There is no reliable experimental data on diffusion of Ni in pyroxene [e.g., 2]. However, the weak correlations between  $^{60}\text{Ni}$  excesses and Fe/Ni ratios in chondrules with evidence for  $^{60}\text{Fe}$  and the lack of evidence for  $^{60}\text{Fe}$  in most chondrules suggest that Ni may be mobile in both olivine and pyroxene under the thermal conditions experienced by type 3.0-3.2 chondrites.

One possible explanation for the two chondrules with high inferred  $(^{60}\text{Fe}/^{56}\text{Fe})_0$  ratios is that they preserve evidence for a relatively high initial ratio that was disturbed in these chondrules and erased by later heating in most objects [4,12]. In order for internal isochrons to be disturbed or reset, it is necessary to redistribute the Fe and Ni isotopes between phases within a chondrule. If Fe moves out of a mineral while

Ni remains behind, an “unsupported” excess of radiogenic  $^{60}\text{Ni}$  might be present. This does not appear to be the explanation for the Semarkona and Krymka chondrules with resolved initial ratios because their isochrons contain spots with very high Fe/Ni ratios (i.e.,  $^{56}\text{Fe}/^{62}\text{Ni}$  ratios of up to  $2 \times 10^5$ ). Alternatively, if radiogenic Ni moves from a phase that originally had a very high Fe/Ni ratio and is concentrated locally (e.g., in sulfide grains), the “unsupported”  $^{60}\text{Ni}$  could give a high inferred initial ratio. A SIMS measurement that includes both phases, could give either a higher or lower Fe/Ni ratio than the original phase.

The Semarkona and Krymka chondrules with resolved initial ratios are radiating pyroxene chondrules, which are peppered with tiny sulfide grains. If most of the Ni has moved from the pyroxenes to the sulfide grains, different Fe/Ni ratios would result from different amounts of sulfide in a measurement volume, but the  $^{60}\text{Ni}$  excess may not vary significantly. If some of the original Ni-rich sulfide (i.e., sulfide not contaminated with Ni from pyroxene) is also present, the data might begin to look like an isochron, but with a poor correlation between excess  $^{60}\text{Ni}$  and Fe/Ni ratio. This is what we observe (Fig. 1 in [4]).

This model requires that the  $(^{60}\text{Fe}/^{56}\text{Fe})_0$  ratio in the region where UOC chondrules formed was relatively high, perhaps  $1 \times 10^{-7}$  or even higher. However, measurements of bulk chondrules give uniformly low  $(^{60}\text{Fe}/^{56}\text{Fe})_0$  ratios [12-14]. For the model to be correct, Fe and Ni must have been redistributed between chondrules and matrix, but there is little direct evidence of this. Additional work will be required to understand the mobility of Ni in UOCs. The degree to which the Fe-Ni system has remained closed in other meteoritic materials must also be investigated.

**References:** [1] Tachibana S. and Huss G.R. (2003) *Ap. J.* 588, L41-L44. [2] Tachibana S. et al. (2006) *Ap. J.* 639, L87-L90. [3] Mostefauoi S. et al. (2005) *Ap. J.* 625, 271-277. [4] Telus et al. (2012) *LPSC XLIII*, #2733 [5] Mishra R.K. and Chaussidon M. (2012) *MAPS*. 47, #5194. [6] Shukolyukov A. and Lugmair G.W. (1993) *Science* 259, 1138-1142. [7] Hester J.J. and Desch S.J. (2005) In *Chondrites and the Protoplanetary Disk*, pp. 107-130. [8] Ogliore R.C. et al. (2011). *Nucl. Instrum. Methods. Phys. Res. B* 269, 1910-1918. [9] Telus M. et al. (2012) *MAPS*. doi: 10.1111/maps.12041. [10] Grossman J.N. and Brearley A.J. (2005) *MAPS*. 40, 87-122. [11] Bonal L. et al. 2006. *GCA*. 70, 1849-1863. [12] Chen et al. (2013) *LPSC XLIV* #2649. [13] Spivak-Birndorf L.J. et al. (2012) *MAPS*. 47, #5365. [14] Tang H. and Dauphas N. (2012) *EPSL*. 359, 248-263. [15] Alexander C.M.O'D. et al. (1989) *GCA* 53, 3045-3057. [16] Huss G.R. and Lewis R. (1994) *Meteoritics* 29, 811-829. [17] Chakroborty S. (2010) *Rev. Mineral. & Geochem.* 72, 603-639. [18] Huss et al. (2006) *MESS II*. p. 567-586. Supported by NASA grants NESSF11 to MT and NNX11AG78G to GRH.