REEVALUATING OUR UNDERSTANDING OF THE ^{60}Fe-^{60}Ni SYSTEM IN CHONDRITES.
M. Telus[1], G. R. Huss[1], K. Nagashima[1], R. C. Ogliore[1] and S. Tachibana[2] [1]HIGP, Univ. of Hawaiʻi at Mānoa, Honolulu, HI 96822 telus@higp.hawaii.edu, [2]Dept. of Earth and Planetary Science, Univ. of Tokyo, Tokyo, Japan.

Introduction: The ^{60}Fe-^{60}Ni system has attracted considerable interest in the last few years because ^{60}Fe ($t_{1/2}$ = 2.6 Myr) is only produced in significant quantities in a stellar source. Its inferred presence in the early solar system has been used as evidence that a supernova was involved in the formation of the solar system [1, 2]. Much of the evidence for high levels of ^{60}Fe in the early solar system has come from ion microprobe studies [e.g., 3-5]. Recently, ICPMS and TIMS work, primarily on achondrites, have not reproduced these high abundances. At the same time, we (re)discovered that the methods that we and many other mass spectrometry groups use to calculate isotope ratios has an intrinsic positive bias [6]. This bias correlates inversely with the number of the counts of the normalizing isotope and can produce apparent correlations that look very much like isochrons.

In order to sort out the confusion generated by the newly rediscovered statistical bias, we have reanalyzed much of the data published over the last few years. In addition, we have measured some new chondrules from unequilibrated ordinary chondrites (UOCs) and remeasured chondrules measured previously. In this abstract, we summarize our findings regarding previously published data. Some of the originally reported detections have gone away, but not all of them. We review the remaining positive results and our new measurements and attempt to summarize the current state of ion probe measurements of the Fe-Ni system.

Reanalyzing Old Data: We recalculated the results reported in [3, 4] using the original ion probe data. We treated the data exactly the way they were treated originally. The only difference was that we calculated the isotope ratios using total counts rather than the means of the individual ratios from the cycle data to reduce the statistical bias.

SIMS Protocol: Iron and Ni isotopes were measured using the Cameca ims 1280 ion microprobe at the University of Hawaiʻi. The data discussed here were measured with several protocols. Our current and most reliable protocol involves measuring Fe and Ni isotopes as positive ions using a 6-8 nA $^{16}O^-$ beam with a 15µm raster. Iron and Ni isotopes are measured simultaneously, with ^{60}Ni, ^{61}Ni, and ^{62}Ni on electron multipliers (EMs) and ^{56}Fe on a Faraday cup. A mass resolving power (MRP) of ~4800 was used. Potential molecular interferences on the Ni isotopes are monitored and tail corrections are made when necessary. Isotope ratios are calculated from the total counts acquired for each isotope during the run. Corrections are applied for detector drift, background, deadtime, and interferences. Instrumental mass fractionation is corrected internally on each individual measurement.

Table 1. Recalculated chondrule and troilite data from [3, 4].

Sample	^{60}Fe/^{56}Fe$_{Pub}$ (×10^{-7})	**^{60}Fe/^{56}Fe$_{tc}$ (×10^{-7})**	^{61}Ni cts/cyc	^{62}Ni cts/cyc
SMK1-4 [4]	2.7 ± 0.8	**1.2 ± 0.9**	~27	~29
SMK2-1 [4]	2.2 ± 1.0	**-1.2 ± 1.4**	~11	~12
SMK2-4 [4]	2.8 ± 2.1	**0.3 ± 2.5**	~41	~44
BIS21 [4]	3.7 ± 1.9	**-1.0 ± 2.6**	~11	~11
BishTr41 [3]	1.1 ± 0.3	**0.4 ± 0.3**	~13	~15
BishTr2 [3]	1.0 ± 0.5	**-0.1 ± 0.4**	~52	~54
BishTr47 [3]	1.3 ± 0.6	**0.1 ± 5.1**	~65	~67
KrymTr1 [3]	1.8 ± 0.7	**-0.2 ± 1.0**	~11	~11
KrymTr12 [3]	1.6 ± 0.8	**-0.1 ± 1.0**	~10	~10

Results: The recalculated results from [3, 4] are presented in Table 1. We find that the published initial ^{60}Fe/^{56}Fe ratios, (^{60}Fe/^{56}Fe)$_{Pub}$, of 1-4×10^{-7}are almost entirely due to the positive statistical bias from reducing the data using the mean of the ratios. When the data are reduced using total counts, (^{60}Fe/^{56}Fe)$_{tc}$, all the initial ratios decrease substantially (Table 1). Initial ^{60}Fe/^{56}Fe ratios for the troilites from Bishunpur and Krymka [3] are all now unresolved from zero. The initial ratios for the Fe-rich silicates from Semarkona and Bishunpur [4] are also mostly unresolved from zero, except for Semarkona sample SMK 1-4, which gives an initial ratio of (1.2 ± 0.9)×10^{-7}. The counts per cycle for ^{61}Ni and ^{62}Ni from the measurements with the highest Fe/Ni ratio are also given in Table 1. The biases in the published data are consistent with the predictions from [6]. All reported errors are 2σ uncertainty.

The (^{60}Fe/^{56}Fe)$_0$ for chondrules from the ordinary chondrites Semarkona (LL3.0), Bishunpur (LL3.15)

Table 2. Preliminary results for the initial (^{60}Fe/^{56}Fe)$_0$ of recently measured chondrules from UOCs.

Sample Name	**Max δ^{60}Ni (‰)**	**^{60}Fe/^{56}Fe$_{tc}$ (×10^{-7})**	χ_ν^2
Semk DAP-1	20 ± 9	2.9 ± 1.4	8
Semk DAP-2		< 0.6	
Bish BM23_s1	11 ± 10	< 2.3	
Bish BM23_13		< 0.7	
Bish BM23_9		< 1.2	
Bish BM23_12		< 0.2	
Bish BM80_13	17 ± 9	2.2 ± 1.1	8
Bish BM80_37		< 2.3	
Kry K3-4 Ch 1		< 0.3	
Kry K2-3 Ch 11	10 ± 6	2.4 ± 0.5	5
Que 11_F		< 1.5	
Que 11_C		< 0.6	
Que 13_6		< 1.0	

Krymka (LL3.2) and QUE97008 (LL3.05) measured within the past year are reported in Table 2. Most chondrules have $(^{60}Fe/^{56}Fe)_0$ ratios that are unresolved from zero. Only a few chondrules seem to show resolved initial ratios, and the regressions for these chondrules have high reduced Chi-squares values (Table 2). For several of them, individual data points exhibit resolved excesses of ^{60}Ni. The largest anomalies are listed in Table 2. They are well-resolved, implying that the effects are real. But the lack of well-defined isochrons appears to reflect isotopic disturbance.

Discussion: Only a few of the pyroxene-bearing chondrules that we have measured show evidence of ^{60}Fe. Although the data do not give good isochrons, they still provide constraints on the initial ^{60}Fe/^{56}Fe at the time the chondrules formed. For instance, Semarkona DAP-1 (Fig. 1), has been measured on several occasions. It consistenly gives an apparent initial ratio of ~(2-3)×10^{-7}, but the linear correlation between ^{56}Fe/^{61}Ni and ^{60}Ni/^{61}Ni ratio is weak, as shown by reduced Chi-square (Table 2). This chondrule has a bleached rim, formation of which may have affected the Fe-Ni system. Regardless, most individual pyroxenes show excesses of ^{60}Ni relative to the standards, with a mean δ^{60}Ni of ~13‰ (Fig. 1). The variable excesses of ^{60}Ni do not represent pre-existing Ni isotopic anomalies because melting and crystallization during chondrule formation would have homogenized the Ni.

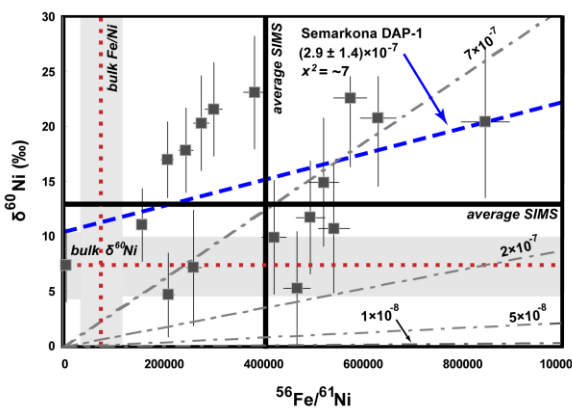

Figure 1. The δ^{60}Ni vs. ^{56}Fe/^{61}Ni isochron plot for Semarkona DAP-1 compared to possible initial (^{60}Fe/^{56}Fe) values. The isochron slope was calculated from DAP-1 data only.

We clearly cannot use DAP-1 to get a precise value for the initial ^{60}Fe/^{56}Fe ratio for the solar system. However, we can use it to provide some constraints on the initial ^{60}Fe/^{56}Fe ratio for the chondrule. The presence of ^{60}Ni excesses of ~5‰ to ~20‰ in the pyroxene of DAP-1 constrains the initial ^{60}Fe/^{56}Fe ratio to be significantly greater than 1×10^{-9} or 1×10^{-8}, numbers that have been suggested for the $(^{60}Fe/^{56}Fe)_{SS}$ based on data from achondrites [7, 8]. If DAP-1 formed with an initial ratio of 1×10^{-8}, the ^{56}Fe/^{61}Ni ratios of DAP-1 pyroxenes would have to be ~10×10^{6} to 46×10^{6}; the latter is >10× higher than the highest ratio observed in this chondrule. An initial ratio of ~5×10^{-8} is probably the minimum that is consistent with the data for DAP-1.

The maximum permissible initial ratio is harder to constrain. Using the average of measured pyroxene values, shown by the heavy lines in Fig. 1, we would infer an initial ratio of 7×10^{-7}, if the initial Ni composition was the same as in our standards. This number is considerably higher than those inferred for other chondrules (Table 2). We estimated the bulk ^{56}Fe/^{61}Ni ratio of DAP-1 (vertical dotted line, Fig. 1) by measuring the Fe and Ni compostion of silicate and sulfide phases using WDS on the JEOL *Hyperprobe* JXA-8500F electon microprobe at UH. A bulk δ^{60}Ni (horizontal dotted line, Fig. 1) was also estimated from our measurements, using the average δ^{60}Ni of pyroxene for silicates and the measured value for sulfide, which plots on the Y-axis of Fig. 1. We estimated the modal abundances with *ImageJ*, an image processing program. Taken at face value, these bulk values would imply an initial ^{60}Fe/^{56}Fe ratio of ~2×10^{-6}, which we consider unlikely. Our estimated δ^{60}Ni is almost certainly too high. Alternatively, DAP-1 may have inherited a bulk ^{60}Ni excess of several permil from it precursors. Either scenario would lower our estimate of the maximum initial ratio, but would not eliminate the evidence for live ^{60}Fe in DAP-1. The major portion of DAP-1 has been set aside for bulk Ni-isotope measurements.

Conclusion: With the present data, it is not possible to get a definitive estimate of the initial ^{60}Fe/^{56}Fe ratio in the early solar system from ion probe measurements. It is clear that the initial ratios reported earlier must be re-evaluated. Apparently, even the least metamorphosed ordinary chondrites are not preserving a high-fidelity record of ^{60}Fe in the early solar system. However, it also seems clear that very low ratios, such as those being reported for various achondrites [e.g., 7, 8], are incompatible with the our data. Assuming all measurements are correct, there are two possible explanations for the observations: 1) few if any of the samples that have been measured to date have avoided isotopic disturbance; they do not preserve their initial Fe-Ni composition, or 2) ^{60}Fe was grossly heterogeneously distributed in the early solar system.

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