

SYNCHROTRON XRF MAPPING OF Fe, Ni AND OTHER ELEMENTS IN UOC CHONDRULES: IMPLICATIONS FOR INTERPRETING ^{60}Fe - ^{60}Ni DATA. M. Telus¹, G. R. Huss¹, R. C. Ogliore¹, K. Nagashima¹, A. Tomkins², ¹HIGP, University of Hawai'i at Mānoa, USA. Email: telus@higp.hawaii.edu. ²School of Geosciences, Monash University, Australia.

Introduction: Iron-60 (half-life = 2.6 Myr) is an extinct short-lived radionuclide. Its presence in the early solar system is inferred through excesses in ^{60}Ni that correlate with the Fe/Ni ratio in various meteorites [e.g., 1]. A well-constrained initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratio in the solar system, $(^{60}\text{Fe}/^{56}\text{Fe})_{\text{SS}}$, is necessary to use the ^{60}Fe - ^{60}Ni system for early solar system chronology. To constrain $(^{60}\text{Fe}/^{56}\text{Fe})_{\text{SS}}$, we have measured *in situ* the Fe-Ni isotopic composition of chondrule ferromagnesian silicates in unequilibrated ordinary chondrites (UOCs) using the ion microprobe at the University of Hawai'i. We found that initial $^{60}\text{Fe}/^{56}\text{Fe}$ ratios [$(^{60}\text{Fe}/^{56}\text{Fe})_0$] for chondrules we have measured during the past few years are mostly unresolved from zero. Only a few chondrules have resolved $(^{60}\text{Fe}/^{56}\text{Fe})_0$ corresponding to $\sim 1 \times 10^{-7}$ [2]. However, the isochron diagrams for these chondrules show weak correlations between excesses in ^{60}Ni and the Fe/Ni ratios. Bulk TIMS and MC-ICPMS measurements of UOC chondrules give $(^{60}\text{Fe}/^{56}\text{Fe})_0 \approx 1 \times 10^{-8}$ [e.g., 1, 3-4], an order of magnitude less than what is inferred from some SIMS data. Both *in situ* and bulk Fe-Ni measurements rely on two assumptions: 1) ^{60}Fe was distributed homogeneously in the solar nebula, and 2) the Fe-Ni system was closed since the event being dated (i.e., chondrule formation).

One explanation for the poorly correlated Fe-Ni ion probe data and the discrepancies between *in situ* and bulk data is that our assumption of closure is incorrect. To evaluate the degree of Fe-Ni redistribution in UOC chondrules, we collected high-resolution X-ray fluorescence (XRF) maps of Fe, Ni and other elements using the Maia detector at the Australian Synchrotron's X-ray fluorescence microscopy (XFM) beamline [5].

Synchrotron measurements: We mapped portions of ~50 chondrules from thin sections of UOCs (Table 1). We used a 12.5 keV incident X-ray beam, which goes all the way through the 30 μm thin sections. We used a spot size of 2.5 μm , dwell times of 31 to 63 ms and a pixel size of 2 μm . The spectra are integrated over the width of the thin section. GeoPIXE spectral data processing software was used to generate quantitative elemental maps of the chondrules [6]. When possible, we confirmed the elemental concentrations derived from GeoPIXE using spot analyses by electron probe at the University of Hawai'i.

Results: Synchrotron XRF maps show clear enrichment of Fe and Ni in many of the cracks in the chon-

drules. Chondrules from all of the meteorites, regardless of petrologic type, show clear evidence for mobilization of Fe and Ni along cracks, including Semarkona, the most pristine chondrite we have available. Iron and Ni mobility is also observed regardless of whether the meteorites are falls or finds (Table 1). Alteration of this kind has not been documented before for chondrites of such low metamorphic grade.

Table 1. Percentage of chondrules from each chondrite that show clear evidence for Fe and Ni mobilization.

Chondrite	Type	Fall/Find	# analyzed	% Fe-Ni mobilization
Semarkona	LL3.00	Fall	16	30
QUE97008	L3.05	Find	7	85
MET96503	L3.10	Find	5	100
Bishunpur	LL3.15	Fall	12	100
Krymka	LL3.20	Fall	9	100

Figure 1 is an Fe, Ni, and Ca distribution map of a portion of a radial pyroxene chondrule with a bleached rim from Krymka (shown in red, green and blue, respectively). Both Fe and Ni are enriched along fractures/veins in the chondrule and in the bleached rim. The black circles are from SIMS measurements, which overlap some of the Fe-Ni enriched cracks and veins. Electron probe spot analyses confirm high Ni concentrations along the cracks and indicate that the phase in the cracks is Fe-Ni oxide and/or Fe-Ni hydroxide. Based on electron probe spot analyses of the glass substrate that the thin section is mounted on, we are confident that the Ni in the cracks is not contamination from the substrate.

Discussion: Contrary to what we have often assumed, our synchrotron XRF maps indicate that Fe and Ni moved into and out of many UOC chondrules, along fractures. This shows that the ^{60}Fe - ^{60}Ni system in chondrules from UOCs was not closed. Instead, Fe and Ni were redistributed between chondrules and matrix as previously suggested by [7]. Mobilization was likely assisted by fluid on the chondrite parent body [8].

These observations provide possible explanations for the discrepancy between bulk and *in situ* Fe-Ni analyses. *In situ* analyses have inferred relatively high $(^{60}\text{Fe}/^{56}\text{Fe})_0$ ratios for a few UOC chondrules [2]. Mild thermal and/or aqueous processing of UOC chondrules

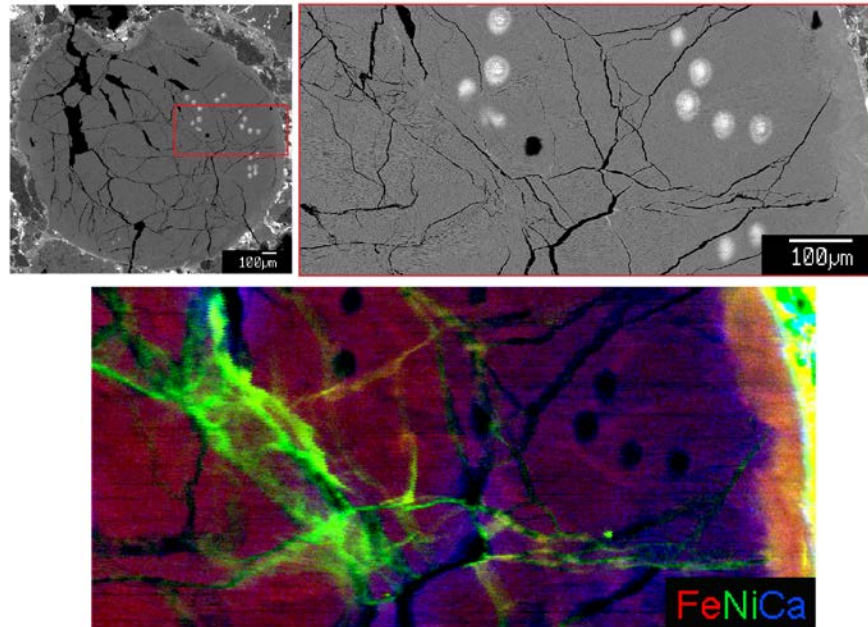


Figure 1. Backscatter electron images (above) and Fe-Ni-Ca X-ray map (below) of a chondrule from Krymka (LL3.2). The elemental map clearly shows Fe and Ni enrichment along the cracks and in the bleached rim of this chondrule. Some ion probe measurements (black circles in the X-ray map) overlap Fe-Ni enriched cracks. The horizontal lines in the X-ray map are an artifact of the scan process.

may have disturbed or completely erased evidence for high initial ratios in chondrules that do not have resolvable ($^{60}\text{Fe}/^{56}\text{Fe}$)₀. This scenario requires that the ($^{60}\text{Fe}/^{56}\text{Fe}$)₀ ratio in the region where these chondrules formed was relatively high, perhaps 1×10^{-7} or higher.

Bulk chondrule Fe-Ni measurements give uniformly low ($^{60}\text{Fe}/^{56}\text{Fe}$)₀ ratios of $\sim 1 \times 10^{-8}$. Redistribution of Fe and Ni between chondrules and matrix could be responsible for these results. If the bulk Ni analyses of the chondrules are dominated by non-radiogenic Ni from the matrix that moved into the chondrules along cracks, measured excess in ^{60}Ni would be lowered, which can significantly affect the ability to resolve excess ^{60}Ni . Bulk chondrule $\epsilon^{60}\text{Ni}$ measurements have 2σ uncertainties of 0.04 – 0.1ε [1, 3]. A chondrule with ($^{60}\text{Fe}/^{56}\text{Fe}$)₀ = 1×10^{-7} and bulk Fe/Ni = 100 should have $\epsilon^{60}\text{Ni}$ of $\sim 0.1\epsilon$. If the bulk Fe/Ni ratio is only 50, as is the case for many chondrules [1], $\epsilon^{60}\text{Ni}$ is reduced to 0.05ε, which is not resolved with these uncertainties. Movement of radiogenic Ni out of a chondrule is likely accompanied movement of non-radiogenic Ni into the chondrule. Loss of radiogenic Ni would lower the measured excesses and further lower the inferred initial ratio. Iron incorporated into chondrules after ^{60}Fe decayed would also lower the inferred initial ratios.

Another possible explanation for the discrepancy between *in situ* and bulk analyses is that, after ^{60}Fe decayed, silicates in some chondrules lost Fe mainly to sulfides and/or metal within the chondrule, with little

to no loss of Ni. If there was no significant loss of Fe to the matrix, this would reduce the Fe/Ni ratio in the silicates, resulting in an apparently high ($^{60}\text{Fe}/^{56}\text{Fe}$)₀ ratio from *in situ* measurements of chondrule silicates. However, to change the inferred ($^{60}\text{Fe}/^{56}\text{Fe}$)₀ ratios from 1×10^{-8} to 1×10^{-7} this way requires that the Fe-silicates lost >90% of the Fe, which is inconsistent with the current silicate compositions.

Conclusions: Synchrotron X-ray data for many UOC chondrules indicate significant exchange of Fe and Ni between chondrules and matrix regardless of petrologic type and whether the chondrites are finds or falls (Table 1, Figure 1). Iron-nickel redistribution on this scale requires caution in collecting and interpreting bulk and *in situ* data for ^{60}Fe - ^{60}Ni chronology.

References: [1] Tang H. and Dauphas N. (2012) *EPSL*. 359, 248-263. [2] Telus M. et al. (2012) *MAPS*. 47, 2013-2030. [3] Chen J. H. et al. (2013) *LPSC XLIV* #2649. [4] Spivak-Birndorf L. J. et al. (2012) *MAPS*. 47, #5365. [5] Paterson D. et al. (2011) *AIP Conf. Proc.* 1365, 219-222. [6] Ryan C. G. (2001) *Nucl. Instr. Meth.* B181, 170-179. [7] Telus M. et al. (2012) *LPSC XLIII*, #2733. [8] Grossman J. N. et al. (2000) *MAPS*. 35, 467-486.

Acknowledgements: This research was undertaken on the XFM beamline at the Australian Synchrotron, Victoria, Australia. Funding from NASA grants NNX11AN62H to MT and NNX11AG78G to GRH. Thanks to K. A. Dyl and J. Cleverley for helpful discussions.