LIVE(?) $^{60}$Fe DURING AQUEOUS ALTERATION OF CHONDRITE PARENT BODIES: EVIDENCE FROM UOCs AND CV CHONDRITES. Patrick H. Donohue$^{1,4}$, Gary R. Huss$^{3}$, Kazuhide Nagashima$^{1}$, and Myriam Telus$^{2}$; $^{1}$Hawai’i Institute of Geophysics and Planetology, University of Hawai’i at Mānoa, Honolulu, Hawai’i 96822, USA (*phd2@hawaii.edu); $^{2}$Department of Terrestrial Magnetism, Carnegie Institute of Washington, 5241 Broad Branch Rd, NW Washington, DC, 20015.

Introduction: Short-lived radionuclides (SLRs) are important chronometers for tracking early solar system evolution. Their initial abundances are evidenced by excesses of daughter products in primitive meteorite components. An initially promising chronometer, $^{60}$Fe-$^{60}$Ni, is difficult to nail down due to mobility in silicates of Fe and the radiogenic daughter, $^{60}$Ni ($^{60}$Ni*) [1]. Iron and Ni mobility increases in the presence of water, which was present in many primitive meteorites. Low-temperature heating also more easily disturbs the Fe-Ni system compared with more robust SLR systems (e.g., $^{26}$Al-$^{26}$Mg). In addition, in 2012 the $^{60}$Fe half-life ($t_{1/2}$) was re-determined, nearly doubling to 2.61 ± 0.04 Myr [2–4]. Despite these challenges, recent findings from in situ analyses support $^{60}$Fe in the early solar system, ($^{60}$Fe/$^{56}$Fe)$_{\text{SLR}}$, at levels between 7.2 × 10^{-8} and 3.8 × 10^{-7} [5,6], though bulk measurements continue to suggest lower values [7].

Here, we have taken a new approach to circumvent insidious Fe and Ni mobilization. Aqueous and thermal alteration processes disrupted primary phases, but also produced new phases, such as fayalite (Fe$_2$SiO$_4$) and magnetite (Fe$_3$O$_4$). In addition to their high Fe content, these phases are relatively pure, with trace Ni content. Thus, they have high Fe/Ni ratios and might be amenable to $^{60}$Fe-$^{60}$Ni dating. If the time interval for formation of these secondary minerals was limited to a few million years after solar system formation, it is plausible the $^{60}$Fe signature would be measurable in the form of positive $^{60}$Ni* anomalies. Given the elapsed time, these measurements will push the limits of detection for in situ analyses.

Samples and Methods: Primary olivine in chondrules may reach $-\text{Fe}_{\text{A0}}$ [Fe$_{\text{A0}}$ = 100*$\text{Fe}/($\text{Fe}+\text{Mg}$)]. Secondary fayalite, found in matrix, generally exceeds $\text{Fe}_{\text{A0}}$ but is small – typically 10-15 µm. Secondary magnetite precipitated from a fluid is nearly pure Fe$_2$O$_4$, whereas that produced by oxidation of metal contains significant Ni [8]. We searched thin sections of 21 meteorites (L, LL, H, and CV) of low petrologic grade (3.0 to 3.4) for secondary fayalite and fluid-deposited magnetite grains. Given the small grain sizes, surveys were manually conducted in backscattered-electron mode. We identified promising magnetite grains in Semarkona (LL3.0) and Kaba (CV3$_{\text{MB}}$), and fayalite grains in Kaba, Vicência (LL3.2), and LEW 86134 (L3.0).

SIMS analyses of magnetite and fayalite were conducted using a Cameca ims 1280 ion microprobe in combined multi-collection jump-scanning mode. The Ni isotopes, $^{60}$Ni, $^{61}$Ni, $^{62}$Ni, were measured on the monocollection electron multiplier along with their closest molecular interferences by jump scanning. $^{60}$Fe was measured on the L1 Faraday cup in multicollection mode at the same field setting as $^{60}$Ni. The mass resolving power for Ni isotopes was ~4800. The primary ion beam was configured in aperture illumination mode to give a ~10 µm spot. The beam current was 1.9 to 3.4 nA. Each spot was pre-sputtered for 1.5 to 3 minutes to eliminate surface contamination. In-house terrestrial magnetite and synthetic fayalite (Fa$_{99}$ [9]) standards were used to determine the relative Fe/Ni sensitivity factor. Excess $^{60}$Ni was calculated using a linear mass fractionation correction ($\Delta^{60}\text{Ni} = \delta^{60}\text{Ni} - 2 \times \delta^{56}\text{Ni}$). The $\delta^{56}\text{Ni}$ and $\delta^{61}\text{Ni}$ ratios were calculate relative to reference values of 7.215 and 0.3136 for $^{60}$Ni/$^{62}$Ni and $^{61}$Ni/$^{62}$Ni, respectively [10].

Results: All determinations of radiogenic $^{60}$Ni were unresolved from zero within 2σ uncertainties, as were the initial $^{56}$Fe/$^{56}$Ni ratios (Fig. 1). We calculated one-sided upper limits (UL) on $^{56}$Fe/$^{56}$Ni ratios, wherein the probability is 95% that the actual ratio falls below the calculated upper limit.

Semarkona. Semarkona magnetite measurements (Fig. 1A) yielded high Fe/Ni ratios similar to [11], with $^{56}$Fe/$^{56}$Ni ratios up to 2.4 × 10^{-7}. At the time of magnetite formation, the initial $^{60}$Fe/$^{56}$Fe, $^{60}$Fe/$^{56}$Fe$_{\text{UL}}$, was <5.9 × 10^{-8}. This is resolvable lower than the (1.1 ± 0.4) × 10^{-7} reported for Semarkona magnetite by [11]. It is likely that their result suffered from ratio bias, which we minimize by calculating ratios from total counts rather than the mean of ratios (cf. [12]).

Kaba. Magnetite and fayalite yielded $^{56}$Fe/$^{56}$Ni ratios up to 3.9 × 10^{-5} and 9.1 × 10^{-5}, respectively (Fig. 1B). Assuming these phases formed simultaneously [13], we regressed all measurements together to yield $^{60}$Fe/$^{56}$Fe$_{\text{UL}}$ of <1.3 × 10^{-7}.

Investigations of Vicência and LEW 86134 are currently underway and will be reported at the meeting.

Discussion: $^{53}$Mn-$^{53}$Cr measurements of fayalite in EET 90161 (L3.05) indicate fayalite formation at ~2.4 Myr after CV CAIs [14,15]. Assuming a similar time interval for Semarkona gives a solar system initial $^{60}$Fe/$^{56}$Fe ratio of <1.1 × 10^{-7}.

Fayalite in CV chondrites has three modes of origin. The large, pure fayalite crystals that we measured formed by precipitation from a fluid [16]. The timing of fayalite formation in Asuka 881317 (CV3) was inferred from $^{53}$Mn-$^{53}$Cr dating to be ~4 Myr after
CAIs [14]. Applying the 4 Myr time interval to our measured ratio of $<1.3 \times 10^{-7}$ gives an estimated upper limit for ($^{60}\text{Fe}/^{56}\text{Fe}_{\text{SS}}$) of $<3.7 \times 10^{-7}$.

These upper limits are within the range inferred for the early solar system from ion microprobe studies of primary phases (Fig. 2) [5,6]. Some anomalous $^{60}\text{Fe}$ signatures (e.g., Semarkona troilite) in Fig. 2 are likely a consequence of Fe and/or Ni mobilization, and potentially analytical ratio bias from SIMS analysis [12,17]. All data on Fig. 2, except magnetite, are from investigations of primary phases or bulk chondrule analyses.

The meteorites that we studied may have experienced a period of heating (up to 300°C in Kaba) subsequent to aqueous processing that formed secondary phases. Extrapolation of diffusivities [18,19 and references therein] to these low temperatures suggest magnetite is the most susceptible to later resetting during this period. The Fe-Ni system in a 30 µm magnetite grain could be fully disturbed within ~1 Myr at 300°C, whereas Mg-rich olivine of the same size would remain viable for >10s of Myr. Diffusion parameters for Ni in pure fayalite are not available, to our knowledge.

Conclusions: The Fe/Ni isotope system is difficult to measure: It is easily disturbed by relatively minor heating; Excesses are difficult to measure due to the high abundance of $^{60}\text{Ni}$; There is limited bulk fractionation of Fe from Ni due to their similar partitioning behavior. These difficulties limit the potential applications of $^{60}\text{Fe}$ as a chronometer. By identifying these issues, we can take steps to avoid potential problems. The early solar system $^{60}\text{Fe}$ abundance is not as high as first thought. The “least altered” meteorites, such as Semarkona, show evidence that Fe and Ni were re-distributed. However, our values for initial solar system $^{60}\text{Fe}$, determined from secondary phases, are not inconsistent with the current consensus [5,6].

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