LIVE(?) ⁶⁰Fe DURING AQUEOUS ALTERATION OF CHONDRITE PARENT BODIES: EVIDENCE

FROM UOCS AND CV CHONDRITES. Patrick H. Donohue^{1*}, Gary R. Huss¹, Kazuhide Nagashima¹, and Myriam Telus²; ¹Hawai'i Institute of Geophysics and Planetology, University of Hawai'i at Mānoa, Honolulu, Hawai'i 96822, USA (*phd2@hawaii.edu); ²Department 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, ⁶⁰Fe-⁶⁰Ni, is difficult to nail down due to mobility in silicates of Fe and the radiogenic daughter, ⁶⁰Ni (⁶⁰Ni^{*}) [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., ²⁶Al-²⁶Mg). In addition, in 2012 the ⁶⁰Fe 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 60Fe in the early solar system, $({}^{60}\text{Fe}/{}^{56}\text{Fe})_{SS}$, 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 (Fe₂SiO₄) and magnetite (Fe₃O₄). 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 ~Fa₄₀ [Fa_% = 100*Fe/(Fe+Mg)]. Secondary fayalite, found in matrix, generally exceeds Fa₇₀ but is small – typically 10-15 μ m. Secondary magnetite precipitated from a fluid is nearly pure Fe₃O₄, 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_{0xB}), 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, 60Ni, 61Ni, 62Ni, were measured on the monocollection electron multiplier along with their closest molecular interferences by jump scanning. ⁵⁶Fe 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 μ m 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 (Fa99 [9]) standards were used to determine the relative Fe/Ni sensitivity factor. Excess ⁶⁰Ni was calculated using a linear mass fractionation correction (Δ^{60} Ni = δ^{60} Ni - 2× δ^{61} Ni). The δ^{60} Ni and δ^{61} Ni ratios were calculate relative to reference values of 7.215 and 0.3136 for 60Ni/62Ni and ⁶¹Ni/⁶²Ni, respectively [10].

Results: All determinations of radiogenic ⁶⁰Ni were unresolved from zero within 2σ uncertainties, as were the initial ⁶⁰Fe/⁵⁶Fe ratios (Fig. 1). We calculated onesided upper limits (UL) on ⁶⁰Fe/⁵⁶Fe 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 ⁵⁶Fe/⁶¹Ni ratios up to 2.4×10^7 . At the time of magnetite formation, the initial ⁶⁰Fe/⁵⁶Fe, (⁶⁰Fe/⁵⁶Fe)₀, was $<5.9 \times 10^{-8}$. This is resolvably lower than the (1.1 ± 0.4) $\times 10^{-7}$ 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 ⁵⁶Fe/⁶¹Ni 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 (⁶⁰Fe/⁵⁶Fe)₀ of <1.3 × 10⁻⁷.

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

Discussion: ⁵³Mn-⁵³Cr 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 ⁵³Mn-⁵³Cr dating to be ~4 Myr after

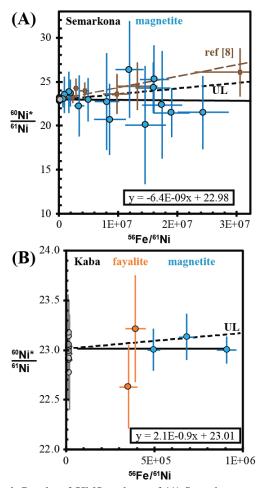


Fig. 1. Results of SIMS analyses of (A) Semarkona magnetite (mgt), compared to [8], and (B) Kaba fayalite (fa) and magnetite. Gray points show the isotopic ratios measured in our terrestrial standards. One-sided upper limits (UL) are discussed in text.

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 Fe/ 56 Fe)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 ⁶⁰Fe 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,

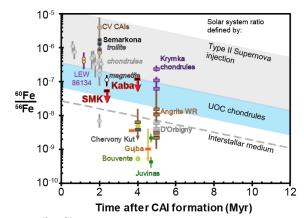


Fig. 2. ⁶⁰Fe/⁵⁶Fe at the time of formation as a function of age. Upper limits from this study for Kaba and Semarkona (SMK), in red, are consistent with previous constraints for UOC chondrules. Data sources: [5,6 and references therein].

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 ⁶⁰Ni; There is limited bulk fractionation of Fe from Ni due to their similar partitioning behavior. These difficulties limit the potential applications of ⁶⁰Fe as a chronometer. By identifying these issues, we can take steps to avoid potential problems.

The early solar system ⁶⁰Fe 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 ⁶⁰Fe, determined from secondary phases, are not inconsistent with the current consensus [5,6].

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